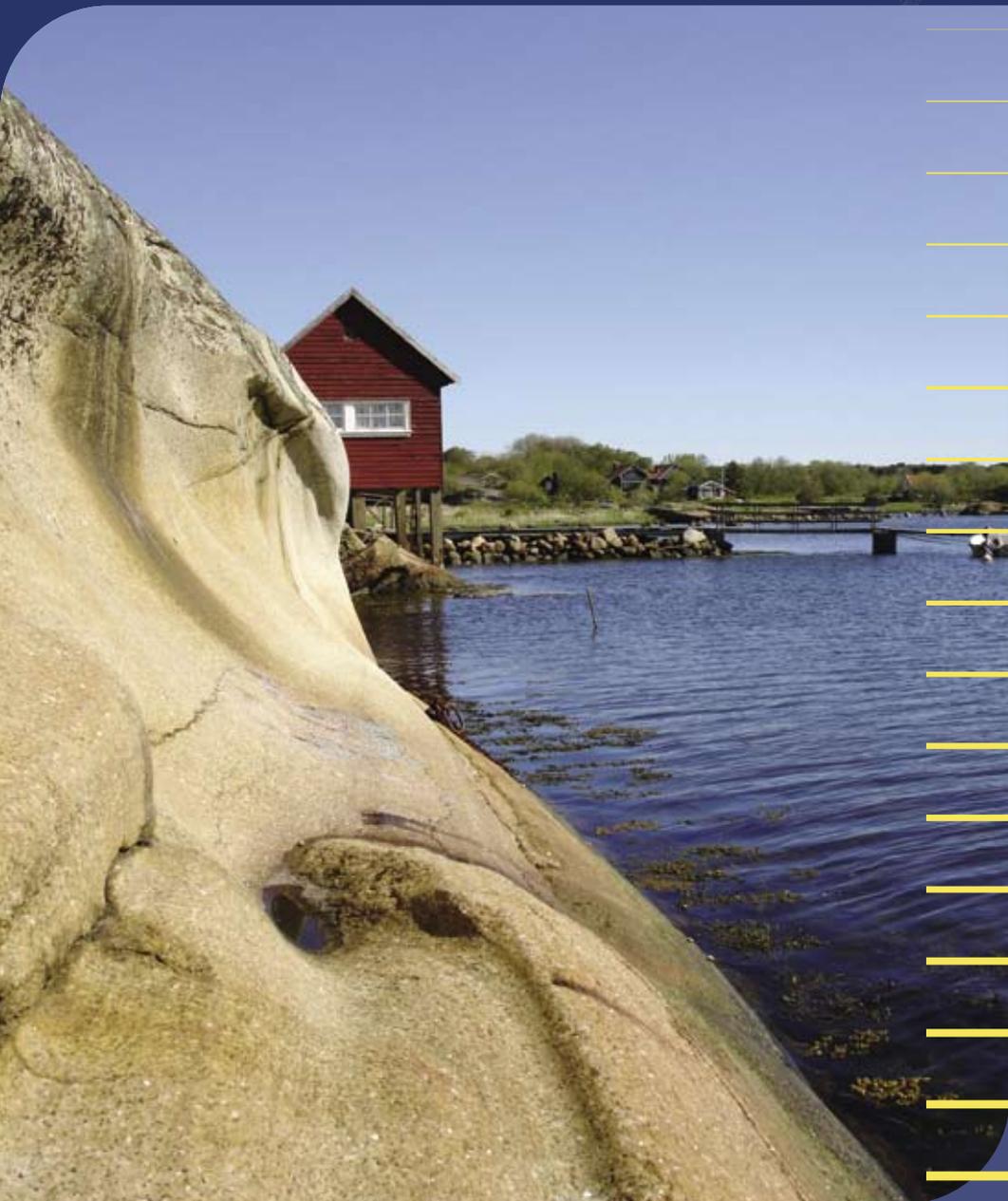


# Radioactivity in the Marine Environment 2004

## Results from the Norwegian Marine Monitoring Programme (RAME)



INSTITUTE OF MARINE RESEARCH



Statens  
strålevern

**Norwegian Radiation  
Protection Authority**

Postboks 55  
N-1332 Østerås  
Norway

*Reference:*

NRPA. Radioactivity in the Marine Environment 2004. Results from the Norwegian National Monitoring Programme (RAME). StrålevernRapport 2006:14. Østerås: Norwegian Radiation Protection Authority, 2006.

*Authors:*

Gäfvert T<sup>1</sup>, Sværen I<sup>2</sup>, Brungot A L<sup>1</sup>, Kolstad A K<sup>1</sup>, Lind B<sup>1</sup>, Gwynn J<sup>1</sup>, Alvestad P<sup>2</sup>, Haldal H E<sup>2</sup>, Føyn L<sup>2</sup>, Strålberg E<sup>3</sup>, Christensen G C<sup>3</sup>, Skipperud L<sup>4</sup>, Salbu B<sup>4</sup>, Drefvelin J<sup>1</sup>, Dowdall M<sup>1</sup>, Rudjord A L<sup>1</sup>

<sup>1</sup>Norwegian Radiation Protection Authority, <sup>2</sup>Institute of Marine Research, <sup>3</sup>Institute of Energy Technology, <sup>4</sup>Norwegian University of Life Sciences

*Key words:*

Radioactivity, marine environment, RAME, monitoring, Norway

*Abstract:*

This report presents results of monitoring of radioactivity in seawater and biota collected along the Norwegian coast and in the North Sea and the Skagerrak in 2004. An overview of discharges from Norwegian sources and data concerning the long-range transport of radionuclides from European nuclear facilities is included.

*Referanse:*

Statens strålevern. Radioaktivitet i det marine miljø. Resultater fra det nasjonale overvåknings-programmet (RAME) 2004. StrålevernRapport 2006:14. Østerås: Statens strålevern, 2006. Språk: engelsk.

*Forfattere:*

Gäfvert T<sup>1</sup>, Sværen I<sup>2</sup>, Brungot A L<sup>1</sup>, Kolstad A K<sup>1</sup>, Lind B<sup>1</sup>, Gwynn J<sup>1</sup>, Alvestad P<sup>2</sup>, Haldal H E<sup>2</sup>, Føyn L<sup>2</sup>, Strålberg E<sup>3</sup>, Christensen G C<sup>3</sup>, Skipperud L<sup>4</sup>, Salbu B<sup>4</sup>, Drefvelin J<sup>1</sup>, Dowdall M<sup>1</sup>, Rudjord A L<sup>1</sup>

<sup>1</sup>Statens strålevern, <sup>2</sup>Havforskningsinstituttet, <sup>3</sup>Institutt for energiteknikk, <sup>4</sup>Universitetet for miljø- og biovitenskap

*Emneord:*

Radioaktivitet, marin miljø, RAME, overvåking, Norge

*Resymé:*

Rapporten inneholder resultater fra overvåkingen av radioaktivitet i sjøvann og biota i 2004 langs norskekysten og i Nordsjøen og Skagerrak. En oversikt over utslipp fra norske kilder og utslippsdata fra europeiske nukleære anlegg som er relevante for langtransport av radioaktivitet til norske havområder er inkludert i rapporten.

Head of project: Anne Liv Rudjord

*Approved:*



Per Strand, Director, Department for Emergency Preparedness and Environmental Radioactivity

49 pages

Published 2006-28-09

Printed number 250 (06-09)

Cover design: Lobo media, Oslo

Printed by Lobo media

*Orders to:*

Norwegian Radiation Protection Authority, P.O. Box 55, N-1332 Østerås, Norway

Telephone +47 67 16 25 00, fax + 47 67 14 74 07

[www.nrpa.no](http://www.nrpa.no)

ISSN 0804-4910

**Radioactivity in the Marine Environment 2004**  
Results from the Norwegian Marine Monitoring Programme (RAME)

**Statens strålevern**  
Norwegian Radiation  
Protection Authority  
Østerås, 2006



# Contents

---

<b>1</b>	<b>Introduction</b>	<b>5</b>
<b>2</b>	<b>Sources of radionuclides in the marine environment</b>	<b>6</b>
2.1	Discharges of anthropogenic radionuclides from Norwegian sources	6
2.1.1	<i>IFE Kjeller and Amersham Health</i>	6
2.1.2	<i>IFE Halden</i>	7
2.2	Unsealed radioactive substances in medical research and industry	8
2.3	Discharge of produced water containing $^{226}\text{Ra}$ and $^{228}\text{Ra}$	9
2.4	Long- range transport of radionuclides	11
2.4.1	<i>Discharge of radionuclides from European nuclear installations</i>	11
2.4.2	<i>Global fallout from nuclear weapons testing</i>	12
2.4.3	<i>Outflow of Chernobyl-contaminated water from the Baltic Sea</i>	13
2.5	Other present and potential sources of radioactivity in the northern marine environment	13
<b>3</b>	<b>Collection of samples</b>	<b>15</b>
3.1	Seawater	16
3.2	Sediment	17
3.3	Seaweed	17
3.4	Fish, crustaceans and molluscs	17
<b>4</b>	<b>Radioactivity in seawater and sediments</b>	<b>18</b>
4.1	Technetium- 99 in seawater	18
4.2	Strontium- 90 in seawater	20
4.3	Caesium- 137 in seawater and sediment	21
4.4	Plutonium- 239+ 240 in seawater	23
4.5	Americium- 241 in seawater	24
4.6	Radium- 226+ 228 in seawater	25
<b>5</b>	<b>Radioactivity in biota</b>	<b>26</b>
5.1	Technetium- 99 in seaweed	26
5.2	Caesium- 137, strontium- 90 and plutonium- 239+ 240 in seaweed	31
5.2.1	<i>Caesium-137 in seaweed</i>	31
5.2.2	<i>Strontium-90 in seaweed</i>	33
5.2.3	<i>Plutonium-239+240 in seaweed</i>	34
5.3	Technetium- 99 and caesium- 137 in crustaceans and molluscs	36
5.4	Caesium- 137 in fish	37
5.4.1	<i>Farmed fish</i>	37
<b>6</b>	<b>Summary and conclusions</b>	<b>40</b>

6.1	Sources	40	
6.2	Radioactivity in seawater and sediment	40	
	6.2.1	<i>Caesium-137 in seawater and sediment</i>	40
	6.2.2	<i>Technetium-99 in seawater</i>	41
	6.2.3	<i>Strontium-90 in seawater</i>	41
	6.2.4	<i>Plutonium-239+240 and americium-241 in seawater</i>	41
	6.2.5	<i>Radium-226 and radium-228 in seawater</i>	41
6.3	Radioactivity in biota	41	
	6.3.1	<i>Technetium-99 in seaweed</i>	41
	6.3.2	<i>Caesium-137 in seaweed</i>	41
	6.3.3	<i>Plutonium-239+240 in seaweed</i>	41
	6.3.4	<i>Technetium-99 and caesium-137 in crustaceans and molluscs</i>	41
	6.3.5	<i>Caesium-137 in fish</i>	42
	<b>References</b>	<b>43</b>	
	<b>Appendix</b>	<b>47</b>	
	Analytical methods	47	
	The Norwegian Radiation Protection Authority (NRPA)	47	
	<i>Determination of <sup>238</sup>Pu, <sup>239+240</sup>Pu and <sup>241</sup>Am activity</i>	47	
	<i>Determination of <sup>99</sup>Tc activity</i>	47	
	<i>Detection of gamma emitters</i>	48	
	The Institute of Marine Research (IMR)	48	
	<i>Detection of gamma emitters</i>	48	
	Institute for Energy Technology (IFE)	48	
	<i>Determination of <sup>99</sup>Tc</i>	48	
	<i>Determination of <sup>90</sup>Sr</i>	49	
	<i>Detection of gamma emitters</i>	49	
	<i>Determination of <sup>239+240</sup>Pu</i>	49	
	Risø National Laboratory	49	
	<i>Determination of <sup>90</sup>Sr in seawater</i>	49	
	<i>Determination of <sup>226</sup>Ra and <sup>228</sup>Ra in seawater</i>	49	
	The Norwegian University of Life Sciences (UMB)	49	
	<i>Determination of <sup>99</sup>Tc</i>	49	

---

# 1 Introduction

The issue of present and potential radioactive contamination in the marine environment has received considerable attention in Norway in recent years. In the late 1980s several accidents and incidents involving nuclear-powered submarines, demonstrated that the risk of the release of radionuclides into the Barents Sea should be considered more carefully. In particular, it became evident that better documentation concerning the radioactivity levels in fish and other seafood was important for the seafood export industries. Furthermore, in the early 1990s, information concerning the dumping of nuclear waste emerged through bilateral environmental cooperation between Norway and Russia. In the years that followed, concern grew regarding the safety of military and civil nuclear installations in the northwest of Russia. This concern was associated not only with possible reactor accidents, but also with the prolonged or sudden release of radionuclides from radioactive waste facilities.

In addition to the potential threats outlined above, radionuclides originating from nuclear weapons fallout, the Chernobyl accident and waste discharged from European reprocessing facilities have been detected in the Norwegian marine environment. In 1994 and 1995, the discharge of  $^{99}\text{Tc}$  from the reprocessing facility at Sellafield in the UK increased sharply, and although this discharge has been reduced slightly, it has continued at a high level up to 2003. The discharge of  $^{99}\text{Tc}$  was substantially reduced from 2004, but it will take 3-4 years before this is observable in Norwegian waters. There has been much public concern about the consequences of such kinds of release, as the radionuclides discharged to the Irish Sea are transported by ocean currents via the North Sea into the Norwegian coastal current and to the Barents Sea. In response to this concern, programmes for the monitoring of radioactivity in the marine environment have been established. Due to the economic importance of the fishing industry and its vulnerability to contamination, as well as any rumours of radio-

active contamination, one of the main objectives of these programmes is to document levels and trends of radionuclides in the Norwegian marine environment.

Other industrial activities, such as mining and oil production, may change the distribution of naturally occurring radionuclides in the environment. The discharge of radium from water produced by oil installations is one area that has recently received special attention.

In Norway there are currently two monitoring programmes concerned with radioactivity in the marine environment, both coordinated by the Norwegian Radiation Protection Authority (NRPA). One is funded by the Ministry of the Environment and focuses on monitoring of radioactivity in the marine environment both in coastal areas and in the open seas, the other by the Ministry of Fisheries which focuses on monitoring of radioactivity in commercially important fish species.

The marine monitoring programme includes the compilation of discharge data from Norwegian sources, in addition to the collection of discharge data relevant for the long-range transport of radionuclides from various sources. Liquid discharge data for 2004 from nuclear installations and recent trends in such discharges are summarised in Chapter 2.

During 2004, samples for monitoring radioactivity in the marine environment were collected in the North Sea, Skagerrak and around the islands of Svalbard, Hopen, Bjørnøya and Jan Mayen. Results from the analysis of these samples are presented in Chapters 4 and 5. In Chapter 6, a summary of the findings and the conclusions is given. In the Appendix, technical information regarding sample preparation techniques and analytical methods employed in the laboratories are presented.

## 2 Sources of radionuclides in the marine environment

This chapter provides updated information and an overview of radionuclide discharges and other sources that are relevant to the Norwegian marine environment. The collection and updating of this information is an integral part of the marine monitoring programme RAME (Radioactivity in the Marine Environment), the main purpose of which is to present recent trends in radionuclide discharges and other sources of radioactivity in the marine environment. Information on discharges from Norwegian sources and on long-range transport of radionuclides from various distant sources is included. Anthropogenic radionuclides are discharged from the nuclear facilities of the Institute for Energy Technology, IFE Kjeller and IFE Halden. Discharges from these facilities are authorised by the NRPA. Unsealed radioactive materials used in medicine and science will primarily be released to the marine environment via sewage treatment plants. The utilisation of such unsealed sources is regulated through guidelines issued by the NRPA.

In recent years, increased attention has been devoted to releases containing elevated levels of naturally occurring radionuclides. Such releases are the result of offshore oil production and, probably, leaching of mine tailings.

The discharge of radionuclides from Norwegian sources is only detectable in the local environment, near the discharge point. The long-range transport of radionuclides originating from fallout from atmospheric nuclear weapons tests (conducted mainly in the 1950s and 1960s), the Chernobyl accident in 1986 and from reprocessing of nuclear fuel are still the main contributors to the general levels of anthropogenic radionuclides found in Norwegian waters.

## 2.1 Discharges of anthropogenic radionuclides from Norwegian sources

### 2.1.1 IFE Kjeller and Amersham Health

IFE Kjeller is located about 20 km east of Oslo. The facilities include a heavy-water-cooled and moderated research reactor with a thermal effect of 2 MW, called JEEP II, a metallurgical laboratory, production facilities for medical radioactive isotopes, and a radioactive waste treatment plant for low-level and intermediate-level waste (LLW and ILW). The liquid LLW originates from these facilities. Liquid effluent is discharged through a designated pipeline to the River Nitelva about 100 km from the sea. The river empties into Lake Øyeren where the water is mixed with the water from the Glomma River. The River Glomma empties into the Oslofjord at the city of Fredrikstad.

A plant manufacturing radiopharmaceutical products, managed by the private company Amersham Health, operates in close collaboration with IFE Kjeller. Authorisation for IFE Kjeller discharges also includes the discharge from this production facility.

**Table 2.1.** Liquid discharges (MBq) from IFE Kjeller (IFE, 2005a; IFE, 2004a; IFE, 2003a; IFE, 2002a; IFE, 2001a), 2000-2004 (including Amersham Health).

Nuclide	2000	2001	2002	2003	2004
<sup>137</sup> Cs	6.7	25.7	25	22	7,9
<sup>134</sup> Cs	0.4	2.5	2.3	1.2	0,35
<sup>131</sup> I	6.3	107	6.0	15	27
<sup>125</sup> I	91	310	350	540	327
<sup>65</sup> Zn	0.9	3.8	<0.4	0.27	0,1
<sup>60</sup> Co	8.7	74	60	55	24
<sup>35</sup> S	142	-	-	-	-
<sup>90</sup> Sr	6.5	1.2	0.6	0.33	2,6
<sup>239+240</sup> Pu	0.2	0.04	0.04	0.034	0,15
<sup>3</sup> H	4.0·10 <sup>5</sup>	1.5·10 <sup>6</sup>	2.4·10 <sup>6</sup>	2.8·10 <sup>6</sup>	2,95·10 <sup>5</sup>

The discharge limit authorised by the NRPA is based on the annual dose to any member of a critical group of the population along the River Nitelva, and shall not exceed 1  $\mu\text{Sv}$ . Each year, IFE Kjeller reports discharge data, the results of their environmental monitoring programme and calculations of effective doses resulting from discharges, to the NRPA. The reported discharges for the period 2000-2004 are summarised in Table 2.1.

**Table 2.2.** Effective doses ( $\mu\text{Sv}$ ) to any member of the critical group from river water exposure pathways as reported by IFE Kjeller (IFE, 2005a; IFE 2004a; IFE, 2003a; IFE 2002a; IFE, 2001a).

2000	2001	2002	2003	2004
0.04	0.21	0.18	0.17	0.07

Effective doses have been calculated by IFE for an individual in the hypothetical critical group exposed through the annual consumption of 20 kg of fish from the river and 100 hours per year presence on the riverbanks. In 2004, the calculated effective dose to this critical group was 0.07  $\mu\text{Sv}$ , corresponding to 7 % of the annual dose limit of 1  $\mu\text{Sv}$ . The effective dose to the critical group varies, as can be seen in Table 2.2, corresponding to between 4 and 21 % of the dose limit, with an average of 13 % for the period 2000-2004.

### 2.1.2 IFE Halden

The Halden Boiling Water Reactor (HBWR) is located in the town of Halden, in the southeast of Norway, close to the Swedish border. The HBWR is heavy-water-cooled and moderated with a thermal effect of 20 MW. The reactor was commissioned in 1959.

IFE Halden is authorised to discharge a limited amount of radioactivity from the research reactor to the River Tista (which empties into the Iddefjord). The discharge limit for liquid waste to the marine environment is based on a dose limit of 1  $\mu\text{Sv}$  per year to a hypothetical critical group, and the actual annual discharge is typically 5-10 % of the authorised limit. Liquid discharges in the period 1999-2003, and the

corresponding doses to the critical group are presented in Tables 2.3 and 2.4.

**Table 2.3.** Liquid discharge (MBq) from IFE Halden, 2000 - 2004 (IFE, 2005b; IFE, 2004b; IFE, 2003b; IFE 2002b; IFE 2001b).

Nuclide	2000	2001	2002	2003	2004
<sup>137</sup> Cs	289	58	110	130	70
<sup>134</sup> Cs	14.5	2	18	8.8	8.0
<sup>131</sup> I	0.4	0.04	0.9	3.6	0.68
<sup>58</sup> Co	13.9	49	5.1	3.3	8.7
<sup>95</sup> Zr	21.4	16	5.4	3.3	4.9
<sup>60</sup> Co	530	440	88	62	68
<sup>125</sup> Sb	116	130	18	0.06	0.002
<sup>124</sup> Sb	1.2	-	0.5	-	-
<sup>95</sup> Nb	38	40	12	8.5	11
<sup>54</sup> Mn	5	7	0.5	0.18	0.73
<sup>51</sup> Cr	610	290	150	130	210
<sup>110m</sup> Ag	3.4	0.5	0.005	0.17	0.001
<sup>144</sup> Ce	15.5	14	5.7	3.2	16
<sup>3</sup> H	$5.2 \cdot 10^5$	$2.4 \cdot 10^5$	$1.1 \cdot 10^6$	$2.7 \cdot 10^5$	$5.4 \cdot 10^5$

The calculation of the effective dose to the critical group is based on the following assumptions:

- An annual consumption of 30 kg of fish from the Iddefjord,
- 200 hours per year exposure on the shore of the fjord,
- 50 hours per year bathing in the fjord, and
- 1000 hours per year boating on the fjord.

**Table 2.4.** Effective doses ( $\mu\text{Sv}$ ) to the critical group from marine exposure pathways as reported by IFE Halden, 2000 - 2004 (IFE, 2005b; IFE, 2004b; IFE, 2003b; IFE 2002b; IFE 2001b).

2000	2001	2002	2003	2004
0.11	0.09	0.02	0.014	0.016

## 2.2 Unsealed radioactive substances in medical research and industry

Unsealed radioactive substances are used in hospitals, research laboratories and various industrial activities. According to regulations from 1981, laboratories etc. handling unsealed radioactive substances must be authorised by the NRPA. With this authorisation, laboratories and hospitals are allowed to discharge activity into the sewage system according to predefined limits. On January 1, 2004, new regulations came into force, describing requirements for handling and discharge of unsealed radioactive sources. At the moment all discharges require authorisation.

Unsealed radioactive substances used in medicine dominate the anthropogenic radioactive discharges to the sewage system. Radioactive solutions are given to patients for diagnostic and therapeutic purposes, and enter the sewage systems mainly by the excretion of urine and faeces.

In some tracer experiments in the offshore oil industry, tritium ( $^3\text{H}$ ) is injected into rock formations or drilled wells, a fraction of which is recovered. Water-based material is discharged to the sea while oil-containing mud must be stored. However, for the consequence assessments required by the NRPA, it is conservatively assumed that all the  $^3\text{H}$  is discharged to the open sea. The release of radionuclides used in research laboratories is in general considered less important, partly because only small quantities of activity are used and partly because some of the waste is collected and sent to the IFE (Kjeller) for storage as radioactive waste. Furthermore, most of these radionuclides have short half-lives and the activity is rapidly reduced during storage. Exact data on the nuclides and amounts discharged from the above sources are not available. However, the manufacturers of unsealed radioactive sources are required to report their sales to NRPA monthly, listing customer, type of radionuclide and quantity (activity). In Table 2.5 the sales of unsealed radioactive sources in Norway in 2004 are

listed. Regarding sales of  $^{99\text{m}}\text{Tc}$ , it is important to consider the short half-life of this radionuclide, as a large fraction will decay before it is even used.

**Table 2.5.** Unsealed radioactive sources (isotopes where total activity exceeds 1 GBq) sold in Norway in 2004.

Nuclide	Half-life	Activity (GBq)
$^{99}\text{Mo}/^{99\text{m}}\text{Tc}$	66 h/6 h	27080
$^{99\text{m}}\text{Tc}$	6 h	4.2
$^{131}\text{I}$	8 d	1889.0
$^{14}\text{C}$	5730 y	1.0
$^{111}\text{In}$	2.8 d	30.0
$^{113\text{m}}\text{In}$	1.66 h	3.7
$^{51}\text{Cr}$	27.7 d	12.9
$^{67}\text{Ga}$	3.3 d	1.3
$^{133}\text{Xe}$	5.3 d	317.7
$^{125}\text{I}$	60 d	19.0
$^{32}\text{P}$	14.3 d	159.9
$^{33}\text{P}$	25.4 d	6.7
$^3\text{H}$	12.3 y	460.9
$^{35}\text{S}$	87 d	80.4
$^{18}\text{F}$	110 min	216.2
$^{123}\text{I}$	13.1 h	90
$^{153}\text{Sm}$	46.7 h	26.0
$^{89}\text{Sr}$	50.6 d	4.6
$^{201}\text{Tl}$	73.1 h	60.7
$^{90}\text{Y}$	64.1 h	17.5

### 2.3 Discharge of produced water containing $^{226}\text{Ra}$ and $^{228}\text{Ra}$

Large volumes of produced water, containing dissolved  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ , are discharged into the sea during oil exploitation. Produced water may be formation water trapped in the reservoirs for millions of years or a mixture of formation water and seawater, when seawater has been injected in order to maintain pressure in the reservoirs during oil and gas production. Although the levels of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  are not technologically enhanced, the natural concentrations of these isotopes in the saline formation water are generally much higher (about 3 orders of magnitude) than the background concentration in seawater (a few mBq per litre). A minor fraction, about 17 % of the total volume produced by Norwegian platforms, is currently reinjected into the reservoirs.

The concentration of radium in the produced water may change over the lifetime of the well. Injection of large volumes of seawater, to maintain the pressure within the well, may lead to dilution of the radium in the produced water, but at the cost of greater water-to-oil ratios later in the production process.

In 2004  $143 \cdot 10^6 \text{ m}^3$  of produced water was discharged to the marine environment from the Norwegian oil and gas industry. The total activity of  $^{226}\text{Ra}$  discharged was about 445 GBq (OLF, 2005). Earlier investigations of radium in produced water from the Norwegian continental shelf have shown that the activity concentration of  $^{228}\text{Ra}$  is approximately equal to the activity concentration of  $^{226}\text{Ra}$  (NRPA, 2005a). In Fig. 2.1 the activity of  $^{226}\text{Ra}$  discharged from Norwegian oil fields are presented. Slightly below 50 % of the total discharged activity of  $^{226}\text{Ra}$  was discharged from the Troll B and C platforms.

There has also been some concern that the use of so called scale inhibitors and scale solvers

may change the behavior of the radium isotopes released to the sea. These compounds are used to prevent precipitation of scale inside the production system and occasionally to dissolve scale in plugged wells and may form complexes with radium ions. If the radium isotopes are discharged in a complexed form they may be more available for marine organisms than if they were released as ions.



Photo: NRPA

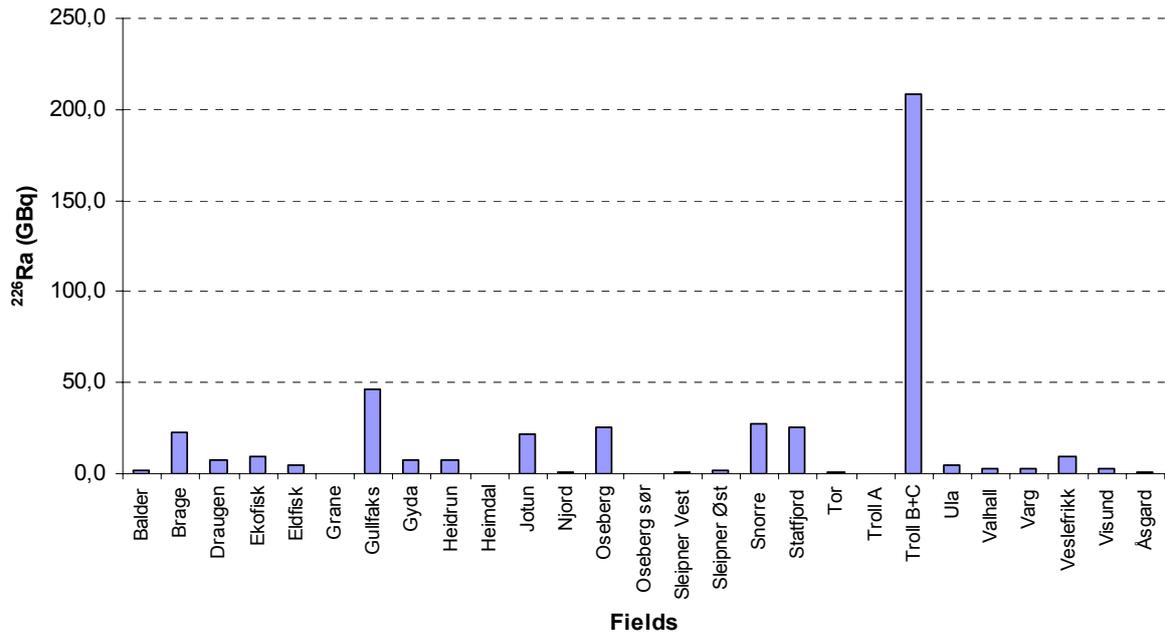
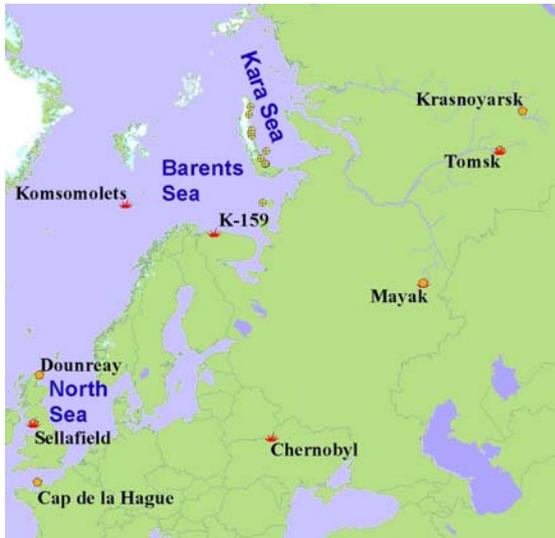


Figure 2.1. Estimated total discharged activity of  $^{226}\text{Ra}$  from Norwegian oil and gas fields in 2004 (OLF, 2005).

## 2.4 Long-range transport of radionuclides

There are several real and potential sources of radioactive contamination of the northeast Atlantic and the Arctic marine environment, as shown in Figure 2.2.

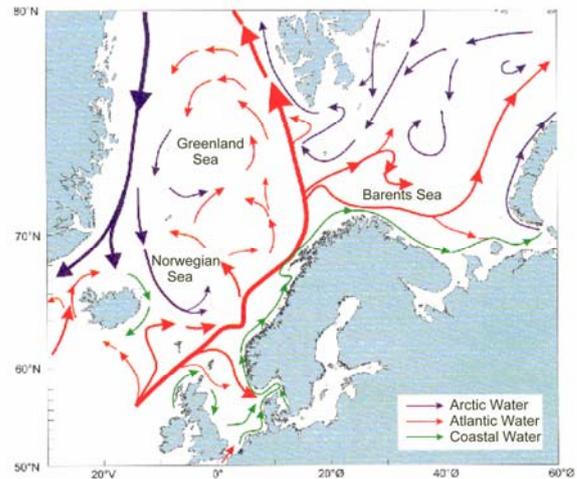


**Figure 2.2.** Sources of radionuclides in the northern marine environment: Chernobyl in the Ukraine, the reprocessing plants at Sellafield, Cap de la Hague and Dounreay, the dumping sites for nuclear waste in the Kara Sea, the sites of the sunken submarines Komsomolets and K-159 and Russian nuclear installations (Mayak, Tomsk and Krasnoyarsk) releasing radionuclides to the Russian rivers Ob and Yenisey.

The primary sources are fallout from atmospheric nuclear weapons testing during the 1950s and 1960s, discharged radionuclides from reprocessing plants and fallout from the Chernobyl accident.

Radioactive contaminants discharged into northern European marine waters can reach Norwegian waters by ocean current transportation. The main ocean currents of the area are presented in Figure 2.3. Some elements, such as Pu and Am, are particle-reactive and will to a large extent sink and end up in the sediments. From the sediments the radionuclides can later be remobilised and transported away by local currents. Remobilised plutonium and  $^{137}\text{Cs}$  from Irish Sea sediments contaminated by previous Sellafield

discharges are one of the main sources of these elements in Norwegian marine waters. Other elements, such as  $^{99}\text{Tc}$  and  $^{90}\text{Sr}$ , are not particle-reactive and will follow currents and can be transported large distances away from the discharge point.



**Figure 2.3.** Overview of the main current system in the North Sea, Norwegian Sea, Greenland Sea and Barents Sea (Aure et al., 1998).

### 2.4.1 Discharge of radionuclides from European nuclear installations

Spent nuclear fuel can be managed in different ways. In a few countries, for instance in France and the UK, spent nuclear fuel is reprocessed. The major part of the waste from these facilities, such as fission products, is stored pending final management and disposal, but a small part is discharged as liquid waste to the marine environment. Discharge limits are set by the relevant national authority.

Sellafield (formerly Windscale) is located on the west coast of Cumbria in the UK. Further information about this reprocessing plant can be found in Amundsen *et al.*, (2003).

The other reprocessing plant in Europe affecting Norwegian marine waters is located at Cap de la Hague in France. Except for tritium ( $^3\text{H}$ ), the discharge of liquid radioactive waste has generally been lower here than from Sellafield.

Springfields is located on the west coast of the United Kingdom, and liquid waste is discharged to the Ribble estuary. This facility is mainly involved in the manufacture of fuel elements for nuclear reactors and the production of uranium hexafluoride. The Dounreay facilities on the northern coast of Scotland were established in 1955, and have mainly been used in the development of technology for fast breeder reactors. Liquid discharge of radioactive waste from Cap de la Hague, Sellafield, Springfields and Dounreay in 2004 is presented in Table 2.6.

**Table 2.6.** Liquid discharge of radionuclides (TBq) from Sellafield, Cap de la Hague, Springfields and Dounreay to the marine environment in 2004 (OSPAR, 2006).

Nuclide	Cap de la Hague	Sellafield	Springfields	Dounreay
$^3\text{H}$	$1.39 \cdot 10^4$	$3.17 \cdot 10^3$	-	0.144
Total- $\alpha$	$1.74 \cdot 10^{-2}$	0.29	0.23	$5.81 \cdot 10^{-4}$
Total- $\beta$	13.1	73.3	116	0.474
$^{14}\text{C}$	8.9	16.3	-	-
$^{60}\text{Co}$	0.26	0.78	-	$2.31 \cdot 10^{-4}$
$^{90}\text{Sr}$	0.14	18.0	-	0.112
$^{99}\text{Tc}$	0.08	14.3	0.12	-
$^{106}\text{Ru}$	6.45	4.42	-	$1.19 \cdot 10^{-3}$
$^{129}\text{I}$	1.37	0.65	-	-
$^{134}\text{Cs}$	$6.4 \cdot 10^{-2}$	0.40	-	-
$^{137}\text{Cs}$	0.79	9.67	-	$2.02 \cdot 10^{-2}$
$^{144}\text{Ce}$	-	0.82	-	$1.06 \cdot 10^{-3}$
Pu- $\alpha$	$6.2 \cdot 10^{-3}$	0.29	-	-
$^{241}\text{Pu}$	0.13	8.1	-	$1.18 \cdot 10^{-4}$
$^{241}\text{Am}$	$2.5 \cdot 10^{-3}$	0.037	-	-
$^{237}\text{Np}$	ND	-	0.0017	-
$^{230}\text{Th}$	-	-	0.11	-
$^{232}\text{Th}$	-	-	$<1.06 \cdot 10^{-3}$	-
U- $\alpha$	-	-	0.046	-
U	$1.7 \cdot 10^{-3}$	436 kg	-	-

In addition to the direct discharges from re-processing, the remobilisation of  $^{137}\text{Cs}$  and plutonium from contaminated sediments in the Irish Sea acts as a secondary source of radionuclides in the marine environment. Substantial

discharges of  $^{238}\text{Pu}$ ,  $^{239+240}\text{Pu}$ ,  $^{241}\text{Am}$  and  $^{137}\text{Cs}$  and other radionuclides in the 1970s and early 1980s resulted in widespread contamination of Irish Sea sediments. It has been shown that these radionuclides can be redissolved and transported out of the Irish Sea. An annual loss of  $1.2 \text{ TBq } ^{239+240}\text{Pu}$  and  $86 \text{ TBq } ^{137}\text{Cs}$  from the Irish Sea has been estimated by Cook *et al.*, (1997), while Leonard *et al.*, (1999) estimated annual losses of plutonium based on remobilisation from surface sediments to be  $0.6 \text{ TBq}$ . The half-lives of plutonium and americium in Irish Sea sediments were estimated to be 58 years and  $\sim 1000$  years, respectively. This implies that remobilisation of radionuclides from Irish Sea sediments is a significant source of anthropogenic radionuclides in the North Sea and Norwegian coastal areas, and will remain so for a long time.

#### 2.4.2 Global fallout from nuclear weapons testing

From a global point of view, atmospheric nuclear weapons tests, conducted between 1945 and 1980, are the largest source of radioactive contamination in the environment. The most intensive test period was between 1952 and 1962, during which the USSR and USA were responsible for most of the tests. The last atmospheric nuclear detonation was conducted in China in 1980. In total, it has been estimated that  $948 \text{ PBq } ^{137}\text{Cs}$ ,  $622 \text{ PBq } ^{90}\text{Sr}$ ,  $6.52 \text{ PBq } ^{239}\text{Pu}$  and  $4.35 \text{ PBq } ^{240}\text{Pu}$  have been released to the environment (UNSCEAR, 2000a), the major part in the northern hemisphere, where most of the tests took place.

Fallout from atmospheric nuclear weapons tests can still be found in the marine environment. About 60 % of the released activity was deposited in the world's oceans. The distribution of the fallout today depends on parameters such as, ocean currents, diffusion and sedimentation rates. Earlier expeditions in the Atlantic have shown that the concentration of  $^{137}\text{Cs}$  is about a few  $\text{Bq m}^{-3}$  (about twice as much as  $^{90}\text{Sr}$ ), between  $45^\circ \text{ S}$  and  $45^\circ \text{ N}$ , with a slight increase to the north (Bourlat *et al.*, 1996). The activity concentration of  $^{239+240}\text{Pu}$

shows the same trend, but the activity concentrations are about 1000 times lower. Reasons for this are that less plutonium was released, but also that plutonium is more particle-reactive and has a higher sedimentation rate, which leads to a shorter residence time in the water.

#### 2.4.3 *Outflow of Chernobyl-contaminated water from the Baltic Sea*

The worst nuclear accident in history occurred on April 26, 1986 in Chernobyl, Ukraine. In the initial explosion and the following fire, about 3-4 % of the nuclide inventory in the core was released to the atmosphere. The major part consisted of volatile radionuclides such as inert gases, iodine isotopes and  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ . Many of the released nuclides had short half-lives and can not be observed in the environment today. The radionuclide that will affect the environment in a long-term perspective is  $^{137}\text{Cs}$ , with a physical half-life of 30 years. In total about 85 PBq of  $^{137}\text{Cs}$  was released to the environment (UNSCEAR, 2000b). The Baltic Sea was the most affected marine area, where about 5 PBq was deposited (Evans, 1991). Shortly after the accident the activity concentration of  $^{137}\text{Cs}$  was extremely high in part of the Baltic Sea. Due to low water exchange and a constant supply from rivers it will take a relatively long time for the levels to decrease. The estimated mean activity concentration of  $^{137}\text{Cs}$  in the Baltic Sea surface water in 2000 was  $50 \pm 20 \text{ Bq m}^{-3}$ , with an effective half-life of  $13 \pm 2$  years (Povinec *et al.*, 2003). Due to the water exchange through the Danish Straits it has been estimated that about 40 TBq flows from the Baltic Sea into the Kattegat annually (Dahlgard, 2002). The  $^{137}\text{Cs}$  will subsequently be transported by the Baltic current which continues as the Norwegian coastal current. This is one of the major sources of  $^{137}\text{Cs}$  in the Norwegian marine environment, together with remobilised  $^{137}\text{Cs}$  from the seabed of the Irish Sea.

## 2.5 Other present and potential sources of radioactivity in the northern marine environment

In addition to discharges from the western European reprocessing plants, northern marine waters have also received an input of various radionuclides from Russian reprocessing plants situated on the tributaries of the Russian rivers Ob and Yenisey (see Figure 2.2). These are the Mayak Production Association in the southern Urals and the Siberian Chemical Combine (Tomsk-7) north of the town of Tomsk, and the Krasnoyarsk Mining and Chemical Combine (KMCC). Uncertainty exists regarding the magnitude of radionuclide contributions from these sources, although estimates have been made. For example, between 1958 and 1993 routine discharges from the KMCC are estimated to have led to an input of 30-100 TBq of  $^{137}\text{Cs}$  into the Kara Sea; a level which is of a similar order of magnitude to inputs from Sellafield-derived  $^{137}\text{Cs}$  to the area (Vakulovsky *et al.*, 1995). However, the fact that the prevailing surface currents tend to transport contamination away from the Kara Sea in predominantly easterly and northerly directions, suggests that the resultant inputs to the Barents Sea has been relatively low. Nonetheless, there is a potential for significant radionuclide contamination following large accidental releases of radioactivity from these nuclear complexes.

Several other sources exist which also represent potential sources of radionuclides in the Barents and Norwegian Seas. Among these are radioactive waste containers dumped in the Barents and Kara Seas by the Former Soviet Union (FSU) and the sunken submarines the Komsomolets and the K-159 in the Norwegian Sea and the Barents Sea. The dumping areas and the locations of the sunken submarines are given in Figure 2.2. Several investigations have been conducted to detect possible leakage from these sources.

As part of the bilateral environmental cooperation with the Russian Federation, a joint Russian-Norwegian expert group has been investigating the condition of the contained dumped objects and has analysed sediment and water samples at these sites.

According to Strand *et al.* (1998), elevated levels of radionuclides in sediments collected in the vicinity of the dumped objects demonstrated that leakage had occurred, but no significant inputs of radionuclides to the open sea from this source have been detected. Following an accident on April 7, 1989, the Russian nuclear submarine the Komsomolets, lies at a depth of 1700 metres, 180 km southwest of Bear Island in the Norwegian Sea. The submarine contains a nuclear reactor and two torpedoes with nuclear warheads. Kolstad (1995) reported low activity concentrations of  $^{134}\text{Cs}$  in seawater samples collected near the submarine, showing that leakage of radiocaesium isotopes may have occurred. Samples of water and sediments collected around the submarine by the Institute of Marine Research and analysed with respect to  $^{137}\text{Cs}$  are presented in Figure 2.4.

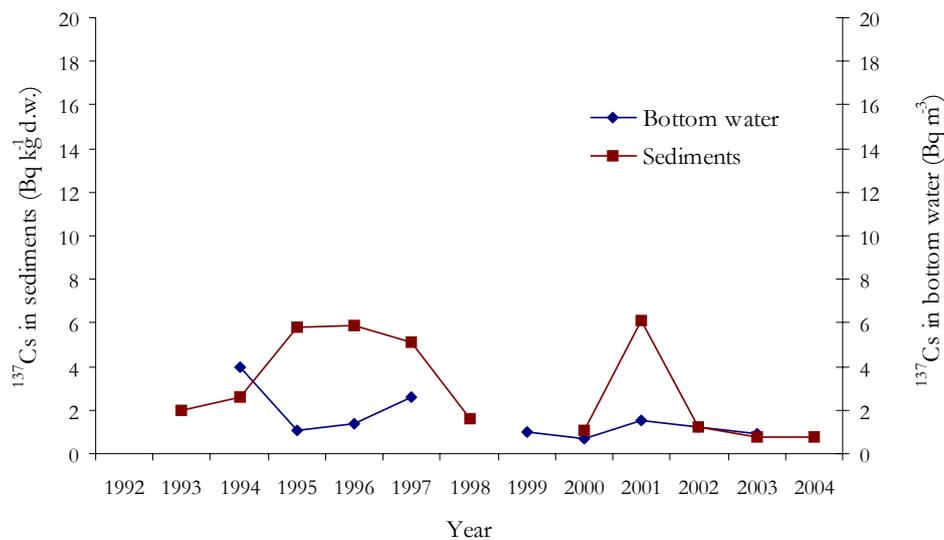


Figure 2.4.  $^{137}\text{Cs}$  in samples from the position of the sunken nuclear submarine "Komsomolets".

### 3 Collection of samples

In 2004 samples were collected in the North Sea, Skagerrak, in selected fjords and at permanent coastal stations including the islands of Svalbard, Jan Mayen, Hopen and Bjørnøya. A geographic overview of the sampling area covered by the marine monitoring programme, with the fixed coastal stations marked, is shown in Figure 3.1.



Figure 3.1. Geographic overview of the sampling area covered by the marine monitoring programmes. (Fixed coastal sampling stations are indicated by yellow dots).

Sampling and analysis were carried out by the Norwegian Radiation Protection Authority (NRPA), the Institute of Marine Research (IMR), the Institute for Energy Technology (IFE) and the Norwegian University of Life Sciences (UMB).

In 2004 samples from the Skagerrak, were collected in the period May 11-24 by the IMR from the vessel R/V "Håkon Mosby". The remaining samples from the North Sea were collected in July by the IMR from the vessel R/V "Johan Hjort" and in November by the NRPA from the research vessel "G.O. Sars" (Fig. 3.2). During the expeditions samples of surface water were collected which were later analysed for  $^{228}\text{Ra}$ ,  $^{226}\text{Ra}$ ,  $^{99}\text{Tc}$ ,  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{241}\text{Am}$  and plutonium isotopes. Sediment was also sampled and later analysed  $^{137}\text{Cs}$ .



Fig. 3.2. R/V G O Sars in Bergen.(Photo: NRPA).

### 3.1 Seawater

For  $^{99}\text{Tc}$ ,  $^{90}\text{Sr}$  and  $^{226}\text{Ra}$  50-100 litres of unfiltered water was collected at each sampling station and later transported to various laboratories for analysis.

For the analysis of  $^{137}\text{Cs}$ ,  $\text{Cu}_2[\text{Fe}(\text{CN})_6]$ -impregnated cotton filters are used as sorbents (Roos *et al.*, 1994). The system consists of a prefilter (1 micron) and two  $\text{Cu}_2[\text{Fe}(\text{CN})_6]$ -impregnated cotton filters connected in series (Fig 3.3). Assuming identical collection efficiency for the  $\text{Cu}_2[\text{Fe}(\text{CN})_6]$ -impregnated cotton filters, the amount of caesium passing through the prefilter can be calculated by considering the two filters as the first two terms in a geometrical series, which has the sum:

$$A(^{137}\text{Cs}) = A_1 / (1 - A_2 / A_1)$$

where  $A_1$  and  $A_2$  are the  $^{137}\text{Cs}$  activities on the first and second impregnated filters, respectively.



Figure 3.3. Filter system used for analysis of  $^{137}\text{Cs}$  in seawater.(Photo: NRPA).

Seawater samples of 200 litres (unfiltered) were collected for the determination of the activity concentrations of  $^{238}\text{Pu}$ ,  $^{239+240}\text{Pu}$  and  $^{241}\text{Am}$ . To collect deep water samples a CTD/Rosette multi bottle sampler was used (Fig 3.4). Plutonium-242 and  $^{243}\text{Am}$  were used as chemical yield determinants. The precipitation of plutonium and americium from the water was performed *in situ* according to the analytical procedure described by Chen *et al.* (1991).



Fig. 3.4. CTD/Rosette multi-bottle sampler.

### 3.2 Sediment

Sediment samples were retrieved from the seabed using a Smøgen box corer. Profiles were collected from the box by slowly inserting PVC tubes into the sediment sample. Surface sediment samples were obtained from the upper 2 cm layer of the sediment remaining in the box-corer.

### 3.3 Seaweed

The NRPA collected seaweed (*Fucus vesiculosus*) samples from eight stations along the Norwegian coastline. At Hillesøy in northern Norway, seaweed is collected every month and at the other locations once per year. In addition, IFE performs monthly or annual seaweed sampling at eleven locations along the coastline, from the Russian border in the north to the Swedish border in the south. Samples of the algae *Laminaria hyperborea* have been collected at two different locations on the Norwegian west coast, Kvitsøy and Steinshamn by the UMB.

### 3.4 Fish, crustaceans and molluscs

NIFES sampled different fish species, crabs and fish from different locations along the Norwegian coastline which were analysed for  $^{137}\text{Cs}$  and  $^{99}\text{Tc}$ . During expeditions to the Barents Sea and the North Sea fish were also collected by the IMR and analysed for  $^{137}\text{Cs}$ . At the request of the NRPA, Labora A/S in Salten was sampling different species of fish and crustaceans from the northern part of Norway.

Lobsters (*Homarus gammarus*) from Arendal have been analysed for  $^{99}\text{Tc}$ . A local fisherman delivered the lobsters to the NRPA immediately after they had been caught.

## 4 Radioactivity in seawater and sediments

### 4.1 Technetium-99 in seawater

Tc-99 has a physical half-life of  $2.13 \cdot 10^5$  years, and originates from the decay of  $^{99}\text{Mo}$ , which is formed either as a fission product from  $^{235}\text{U}$  or  $^{239}\text{Pu}$  or by neutron activation of  $^{98}\text{Mo}$ . The main source of  $^{99}\text{Tc}$  in Norwegian waters is liquid discharge from the reprocessing plant at Sellafield. In oxygenated seawater  $^{99}\text{Tc}$  is present as the highly soluble pertechnetate ion ( $\text{TcO}_4^-$ ). Due to its conservative behaviour in seawater,  $\text{TcO}_4^-$  has the ability to be transported by ocean currents without being significantly affected by sedimentation processes. From the Irish Sea,  $^{99}\text{Tc}$  is transported by ocean currents to the North Sea and via the Norwegian Coastal Current up to the Barents Sea. The transit time (the time between a specific discharge and the maximum activity concentration from that discharge reaching the sampling location) for  $^{99}\text{Tc}$  to reach this area (Hillesøy) from the Irish Sea has been estimated to 3-4 years (Brown *et al.* 2002; Dahlgaard, 1995).

During 2004, seawater samples were collected in the North Sea and in the Skagerrak and analysed for  $^{99}\text{Tc}$ . The results are presented in Figure 4.1. In the North Sea and the Skagerrak the activity concentration of  $^{99}\text{Tc}$  range from 0.11 to  $3.3 \text{ Bq m}^{-3}$ , where the highest levels were found off the coast of Scotland and the lowest levels in Atlantic water. The activity concentrations found in the Norwegian Coastal Current are similar to those observed in 2001 (Gäfvvert *et al.*, 2003). Monthly samples have also been collected at Hillesøy (Fig. 4.2). Average activity concentration in seawater at Hillesøy in 2004 was  $0.82 \text{ Bq m}^{-3}$ , which is similar to what was observed in 2003.

During 2004, seawater samples were collected from coastal sites around the Svalbard archipelago and in the West Spitsbergen Current (WSC). Tc-99 activity concentrations from Svalbard coastal sites (including

monitoring stations on Hopen and Bjørnøya) ranged from 0.09 to  $0.22 \text{ Bq m}^{-3}$  (Fig. 4.3). Tc-99 activity concentrations in the WSC in 2004 were comparable to those observed around Svalbard and where subsurface samples were taken, activity concentrations of  $^{99}\text{Tc}$  were observed to decrease with increasing depth (Fig. 4.3).

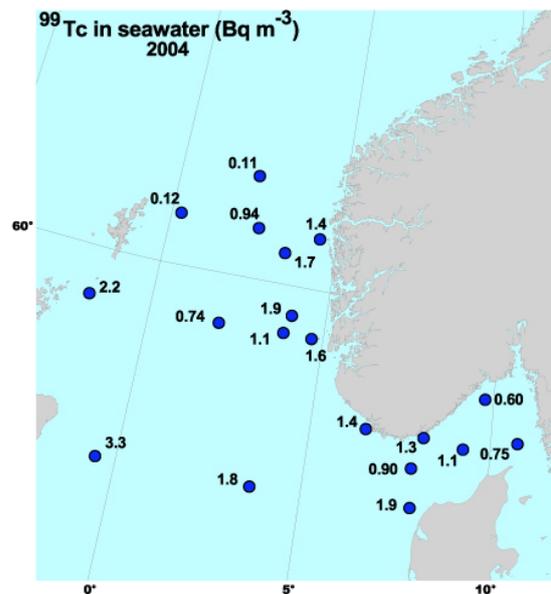


Figure 4.1. Activity concentration ( $\text{Bq m}^{-3}$ ) of  $^{99}\text{Tc}$  in seawater samples collected in the North Sea and Skagerrak in 2004.

Comparison of these values with earlier observations (Gwynn *et al.*, 2004) reveal that seawater activity concentrations for 2004 are generally lower than those reported for same region in 2000 and 2001. However, current activity concentrations of  $^{99}\text{Tc}$  in seawater in the Svalbard region remain above 1994 pre-EARP values of  $0.04 \text{ Bq m}^{-3}$  (Kershaw *et al.*, 1999).

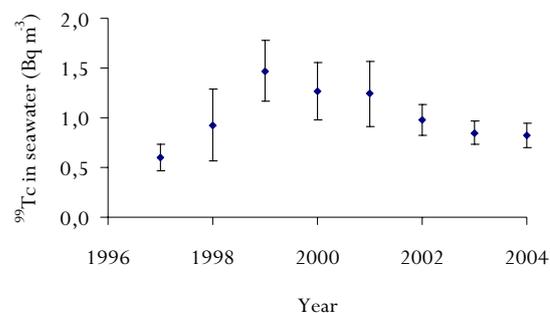


Figure 4.2. Annual average activity concentration ( $\text{Bq m}^{-3}$ ) of  $^{99}\text{Tc}$  in seawater at Hillesøy

Additionally in 2004, samples of seawater from various depths (Fig. 4.3) were taken in a transect across the East Greenland Current (EGC). Activity concentrations in the top 150m of the EGC (Polar Water flowing out of the Arctic Ocean) ranged from 0.11 to 0.16 Bq m<sup>-3</sup>, while two samples taken from 200m (returning Atlantic Intermediate Water) showed comparable activity concentrations of 0.16 ±0.02 and 0.17 ±0.02 Bq m<sup>-3</sup>. Activity concentrations of samples taken from below 800m (recirculating Deep Water) were typically lower than those observed in the top 200m, with the exception of one sample taken from 3000m with an activity concentration of 0.18 ±0.02 Bq m<sup>-3</sup>. Activity concentrations of <sup>99</sup>Tc in seawater at the coastal monitoring station on Jan Mayen in the Greenland Sea for 2004 were lower than those observed in the East Greenland Current ranging from 0.06 to 0.10 Bq m<sup>-3</sup>, but were comparable to values for this station from previous years (NRPA, 2004; NRPA, 2005b).

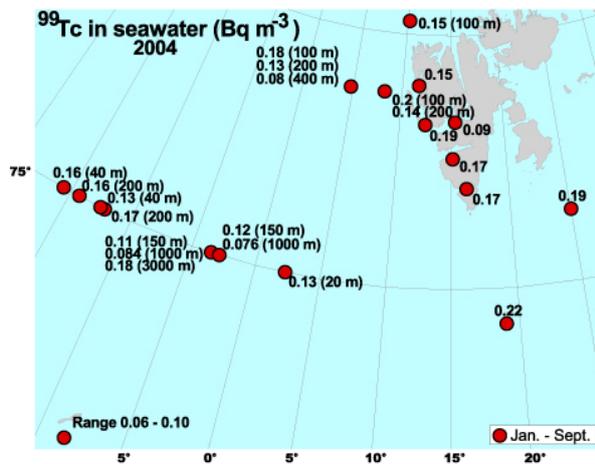
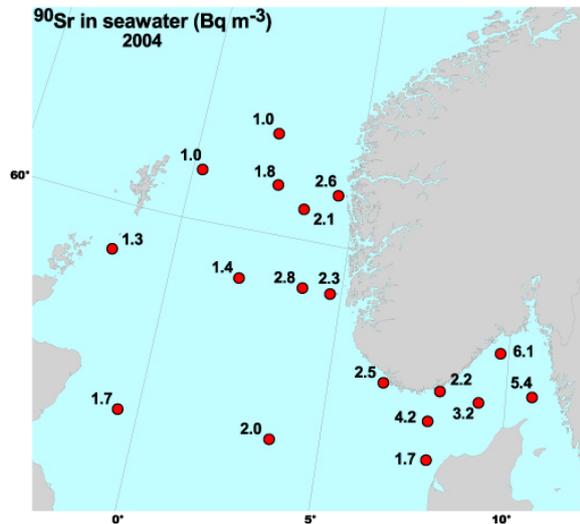


Figure 4.3. Activity concentration (Bq m<sup>-3</sup>) of <sup>99</sup>Tc in seawater samples collected in northern marine waters in 2004.

## 4.2 Strontium-90 in seawater

Strontium-90 is a fission product with a physical half-life of 29 years. Similar to  $^{99}\text{Tc}$ ,  $^{90}\text{Sr}$  is a conservatively behaving element in the marine environment.



**Figure 4.4.** Activity concentration ( $\text{Bq m}^{-3}$ ) of  $^{90}\text{Sr}$  in surface water samples collected in the Norwegian Sea and at coastal stations in 2004.

Seawater has been sampled in the North Sea and the Skagerrak and analysed for  $^{90}\text{Sr}$ . The main sources of  $^{90}\text{Sr}$  in these waters are discharge of liquid waste from reprocessing plants (mainly Sellafield), fallout from atmospheric nuclear weapons tests conducted mainly in the 1950s and 1960s and outflow of water from the Baltic Sea. The annual discharge of  $^{90}\text{Sr}$  from Sellafield between 1998 and 2004 has been in the range of 14 to 31 TBq. The results for 2004 are presented in Figure 4.4 and Table 4.1.

In the northern North Sea and in the Skagerrak, the activity concentration ranged from  $1.0 \text{ Bq m}^{-3}$  to  $6.1 \text{ Bq m}^{-3}$ . The highest concentrations were found off the southern coast of Norway in the Skagerrak. Comparing the activity concentrations observed in 2001 (Gäfvvert *et al.*, 2003) in the North Sea and the Skagerrak one can see that the level of  $^{90}\text{Sr}$  was somewhat higher in 2004 in the Skagerrak, while the levels in the North Sea were similar to those observed in 2001. One explanation for the higher activity concentration of  $^{90}\text{Sr}$  in the Skagerrak could be a large outflow of seawater from the Baltic Sea in

2004. Typical activity of  $^{90}\text{Sr}$  in Baltic seawater have been reported to be in the rang from 8 to  $12 \text{ Bq m}^{-3}$  (Vartti *et al.*, 2006).

**Table 4.1.** Activity concentration of  $^{90}\text{Sr}$  in seawater sampled at coastal stations in 2004.

Location	$^{90}\text{Sr}$ ( $\text{Bq m}^{-3}$ )	Sampling date
Lista	2.8	02.09.2004
Narestø	3.4	01.09.2004
Tjøme	4.1	03.09.2004
Gr. Jakobselv	1.5	01.09.2004

### 4.3 Caesium-137 in seawater and sediment

Caesium-137 is a fission product with a half-life of 30 years. The main sources of  $^{137}\text{Cs}$  in the North Sea and the Skagerrak are fallout from atmospheric nuclear weapons tests in the 1950s and 60s, outflowing water from the Baltic Sea and  $^{137}\text{Cs}$  remobilised from Irish Sea sediments. Runoff from land, from the areas with the highest Chernobyl fallout can also contribute locally in coastal water. Like  $^{99}\text{Tc}$  and  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  is also a conservatively behaving radionuclide in seawater.

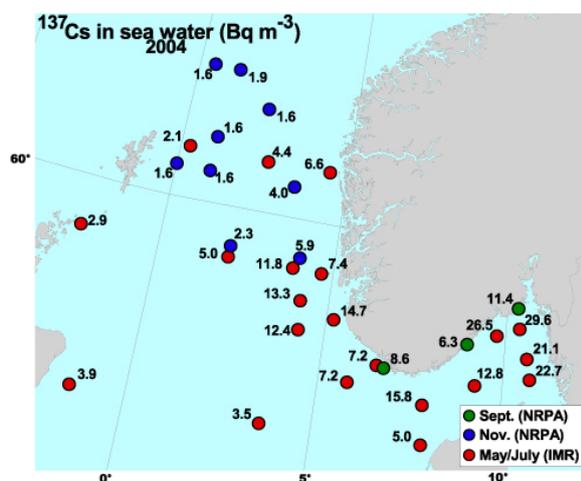


Figure 4.5. Activity concentration ( $\text{Bq m}^{-3}$ ) of  $^{137}\text{Cs}$  in seawater samples collected in the North Sea and in the Skagerrak in 2004.

Observed levels of  $^{137}\text{Cs}$  in the North Sea, the Skagerrak and at the coastal sampling stations at Hillesøy and Grense Jakobselv are presented in Figure 4.5 and Table 4.2.

Table 4.2. Activity concentration of  $^{137}\text{Cs}$  in seawater sampled at Hillesøy and Grense Jakobselv in 2004.

Location	Activity conc. of $^{137}\text{Cs}$ ( $\text{Bq m}^{-3}$ )
Hillesøy	2.4
Gr. Jakobselv	2.8

The activity concentration in surface water ranged from 1.6 to  $29.6 \text{ Bq m}^{-3}$ , where the highest concentrations were in the Skagerrak. The high concentrations found in the Skagerrak and the North Sea in May 2004 is most likely due to a large outflow of Baltic seawater at this

time. In Fig. 4.6 the relation between salinity and activity concentration is shown in the samples collected in May. The highest activity concentration of  $^{137}\text{Cs}$  was found in the samples with the lowest salinity. Typical activity concentration in Baltic seawater is between 30 and  $50 \text{ Bq m}^{-3}$  (Varti *et al.*, 2006).

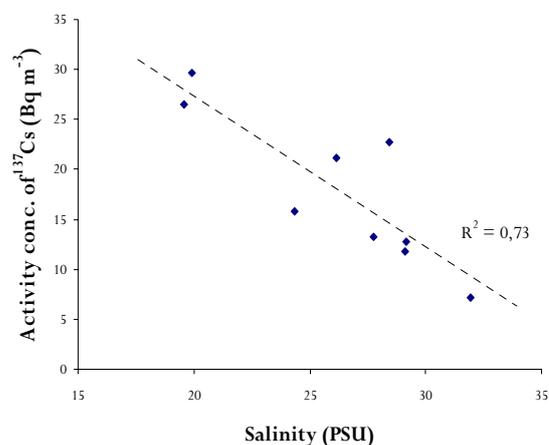


Figure 4.6. Activity concentration of  $^{137}\text{Cs}$  versus salinity in samples collected in May 2004 in Skagerrak and the North Sea.

Cs-137 has also been analysed in surface sediments (upper 2 cm layer) from the northern North Sea and the Skagerrak. The results are presented in Fig. 4.7 and range from 0.2 to  $13.4 \text{ Bq kg}^{-1}$  (d.w.). Highest concentrations were found in the Skagerrak, which can be explained by relatively high sedimentation rate in this area. The results are similar to those found in 2001 in the same area.

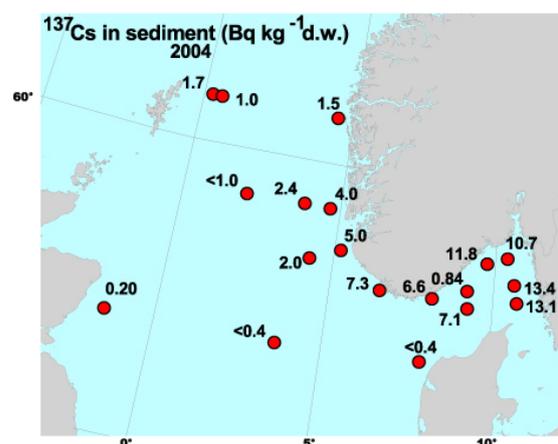
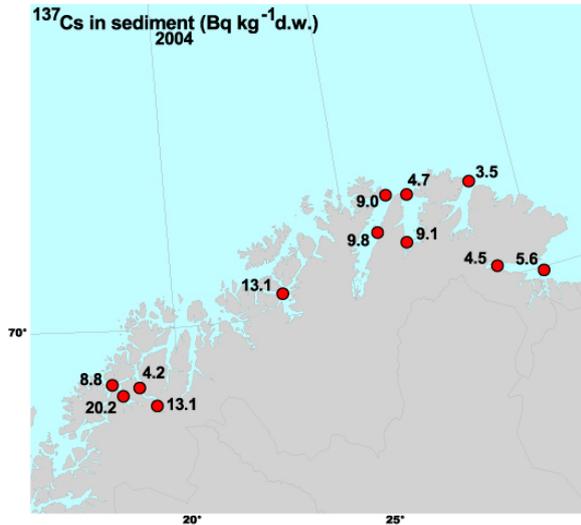


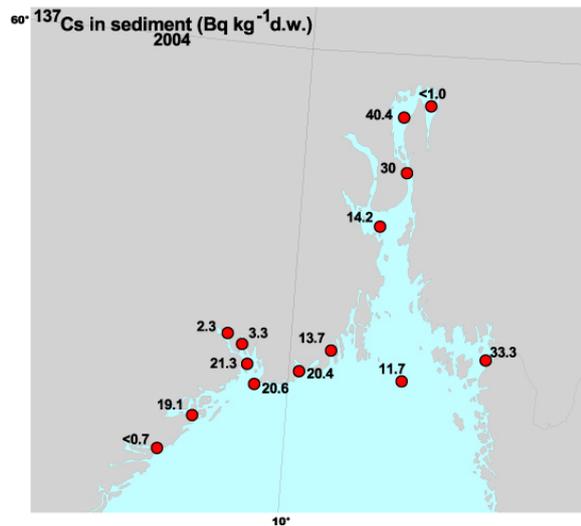
Figure 4.7. Activity concentration ( $\text{Bq kg}^{-1} \text{ d.w.}$ ) in surface sediment from the North Sea and the Skagerrak in 2004.

Surface sediment was also sampled in selected fjords in the northern and southern part of Norway and in the Barents Sea (Fig. 4.8, 4.9 and Fig. 4.10).



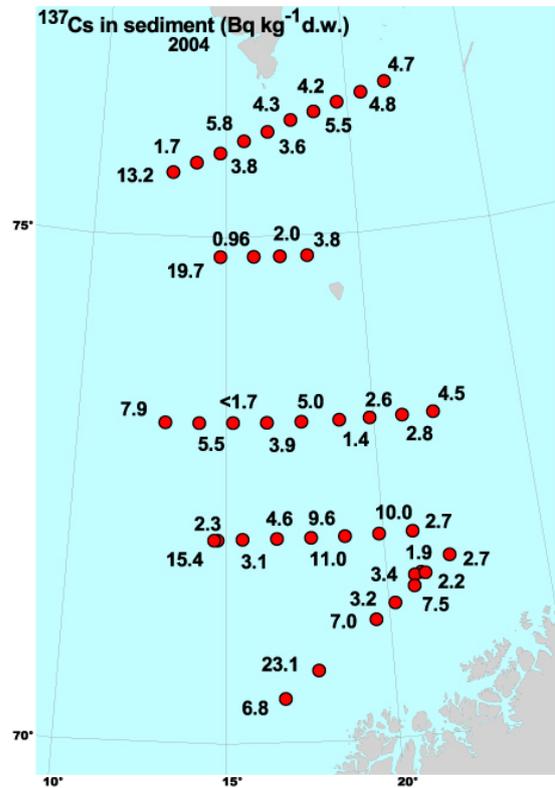
**Figure 4.8 .** Activity concentration ( $Bq\ kg^{-1}\ d.w.$ ) of  $^{137}Cs$  in fjord surface sediment from the northern part of Norway in 2004.

The activity concentration of  $^{137}Cs$  ranged from  $3.5\ Bq\ kg^{-1}\ (d.w.)$  in sediments collected in the northern part of Norway and  $<0.7$  to  $40.4\ Bq\ kg^{-1}\ (d.w.)$  in sediment from fjords in the southern part of Norway. Highest levels were found in fjords in the southern part of Norway.



**Figure 4.9 .** Activity concentration ( $Bq\ kg^{-1}\ d.w.$ ) of  $^{137}Cs$  in fjord surface sediment from the southern part of Norway in 2004.

In the Barents Sea the activity concentration of  $^{137}Cs$  in sediments ranged from  $< 1.7\ Bq\ kg^{-1}\ (d.w.)$  to  $23.1\ Bq\ kg^{-1}\ (d.w.)$ .



**Figure. 4.10.** Activity concentration ( $Bq\ kg^{-1}\ d.w.$ ) of  $^{137}Cs$  in surface sediment from the Barents Sea in 2004.

#### 4.4 Plutonium-239+240 in seawater

Plutonium-239 ( $T_{1/2} = 24\,110$  y) and  $^{240}\text{Pu}$  ( $T_{1/2} = 6\,563$  y) belong to the transuranium elements and are mainly produced by neutron capture (with subsequent beta decay) by  $^{238}\text{U}$  and  $^{239}\text{Pu}$ , respectively. The behavior of plutonium in the marine environment is complex due to its different possible oxidation states. Under oxidizing conditions Pu is generally found in the more soluble +V and +IV state, while in reducing environments the more particle-reactive +III and +IV states dominate. The main source of  $^{239+240}\text{Pu}$  in northern Norwegian marine waters is global fallout from atmospheric nuclear weapons tests in the 1950s and 1960s. Another possible source is remobilised plutonium from Irish Sea sediments.

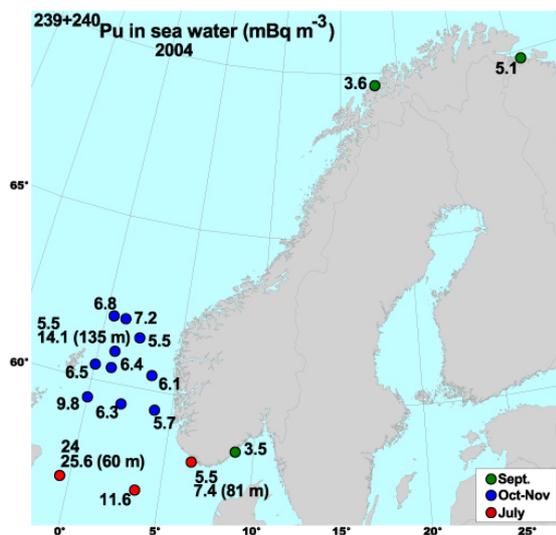


Figure 4.11. Activity concentration ( $\text{mBq m}^{-3}$ ) of  $^{239+240}\text{Pu}$  in surface water samples collected in the North Sea and the Skagerrak and along the coast in 2004.

Observed levels in samples collected in 2004 are presented in Figure 4.11 and range from 3.5 to 25.6  $\text{mBq m}^{-3}$ . The highest level was observed in water collected off the coast of Scotland, showing that Sellafield is a source to plutonium in the North Sea, where part of the  $^{239+240}\text{Pu}$  comes from remobilised plutonium from contaminated Irish Sea sediments. The levels of  $^{239+240}\text{Pu}$  are generally lower than those observed in 2001 (Gäfvvert *et al.*, 2003).

## 4.5 Americium-241 in seawater

Americium-241 belongs to the transuranium elements and has a physical half-life of 432 years. It is produced, for example, in nuclear reactors by successive neutron capture by  $^{239}\text{Pu}$ . Americium-241 is finally formed by beta-decay of  $^{241}\text{Pu}$  ( $T_{1/2} = 14.35\text{ y}$ ). Main sources of  $^{241}\text{Am}$  in the environment are fallout of  $^{241}\text{Pu}$  from nuclear weapon tests in the 1950s and '60s and the discharge of  $^{241}\text{Am}$  and  $^{241}\text{Pu}$  from reprocessing plants.

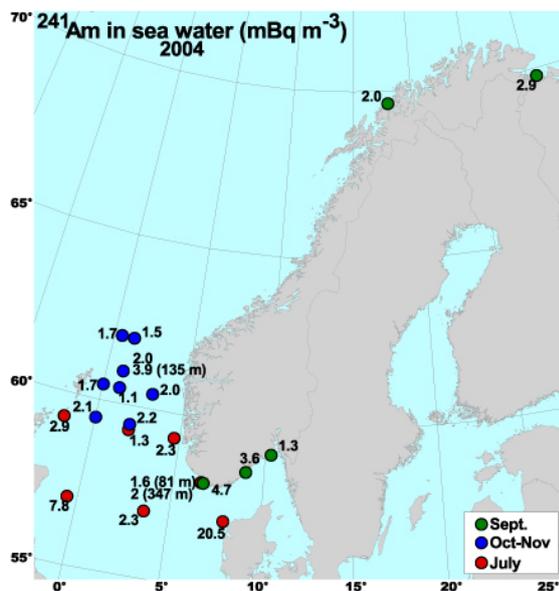


Figure 4.12. Activity concentration ( $\text{mBq m}^{-3}$ ) of  $^{241}\text{Am}$  in seawater from the North Sea and the Skagerrak in 2004.

Measured activity concentrations of  $^{241}\text{Am}$  in the North Sea and the Skagerrak in 2004 are presented in Figure 4.12 and range from 1.1 to  $20.5\text{ mBq m}^{-3}$ . The highest activity concentrations were found off the coast of Denmark and off the coast of Scotland. The  $^{241}\text{Am}$  found in the water column today, can be due to both the present discharge from Sellafield of  $^{241}\text{Am}$  and earlier discharges of  $^{241}\text{Pu}$  (which decays to  $^{241}\text{Am}$ ).

#### 4.6 Radium-226+228 in seawater

Radium-226 is a naturally occurring radionuclide with a physical half-life of 1 600 years. As a member of the natural decay chain starting with  $^{238}\text{U}$  ( $t_{1/2} = 4.47 \cdot 10^9$  years), it is continuously produced by the decay of  $^{230}\text{Th}$ , and can be found in different concentrations in the environment. In the marine environment  $^{226}\text{Ra}$  is naturally supplied from both the sediments (the mother nuclide  $^{230}\text{Th}$  is particle reactive and is mainly found attached to sediment particles) and by river water to the oceans. Typical activity concentration of  $^{226}\text{Ra}$  in Atlantic surface water has been reported to be about  $1.3 \text{ Bq m}^{-3}$  (IAEA, 1990). Radium-228 ( $T_{1/2} = 5.75 \text{ y}$ ) is the daughter of  $^{232}\text{Th}$  and is mainly produced in marine sediments. The highest concentration is found in coastal water where the mixing with oceanic water is limited. An anthropogenic source of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  in the North Sea is produced water from oil reservoirs. Due to the relatively high solubility of radium, produced water can contain more than a thousand times the activity concentration normally found in seawater.

Atlantic water entering the North Sea (Fig 4.13). The slightly higher activity concentration of  $^{226}\text{Ra}$  in the Skagerrak indicates that outflowing seawater from the Baltic Sea contains higher activity concentration of  $^{226}\text{Ra}$  than North Sea water. The  $^{228}\text{Ra}/^{226}\text{Ra}$  ratio varied from 0.1 in Atlantic water to 2.0 in coastal water. This agrees relatively well with earlier reported  $^{228}\text{Ra}/^{226}\text{Ra}$  ratios in Atlantic and coastal water further north (Moore *et al.*, 1980). The levels of  $^{226}\text{Ra}$  found around the Troll B and C platforms were not significantly higher than levels found in coastal water further south. However, at the time of the sampling there was no information available on the exact direction of the discharge plume, and thus these samples may not represent the highest activity concentration that could be found around these platforms.

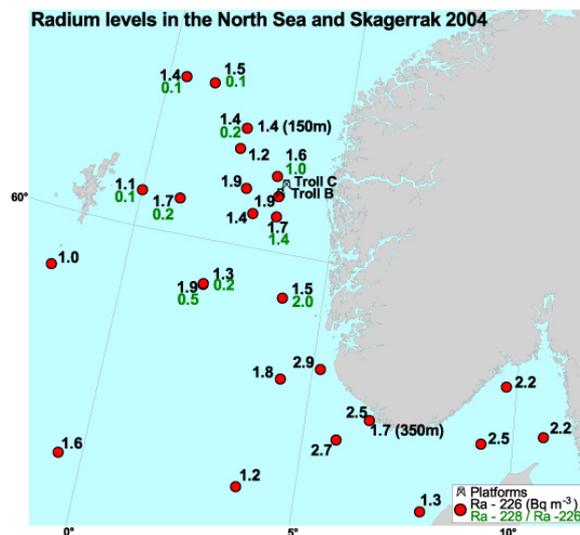


Figure 4.13. Activity concentration ( $\text{Bq m}^{-3}$ ) of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}/^{226}\text{Ra}$  ratio in seawater from the northern North Sea and the Skagerrak 2004.

The activity concentration of  $^{226}\text{Ra}$  observed in the North Sea and the Skagerrak in 2004 ranged from  $1.0$  to  $2.9 \text{ Bq m}^{-3}$ , where the highest levels were found in coastal water and the lowest in

## 5 Radioactivity in biota

### 5.1 Technetium-99 in seaweed



**Figure 5.1.** Brown seaweed (*Fucus vesiculosus*) (Photo:NRPA).

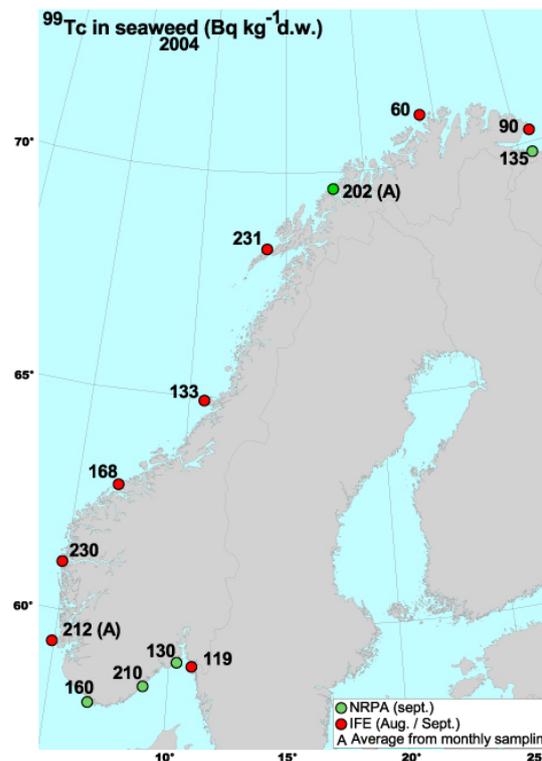
Seaweed is an excellent bioindicator for  $^{99}\text{Tc}$  in the marine environment. It has a very high ability to concentrate  $^{99}\text{Tc}$  from seawater and is easy accessible in most coastal areas. During 2004, seaweed (*Fucus vesiculosus*, Fig 5.1) was collected at the permanent coastal sampling stations along the Norwegian coastline and analysed for  $^{99}\text{Tc}$ . At two locations, Hillesøy and Utsira, sampling was performed monthly. At the remaining sites sampling was conducted in August or September. The results of the analyses are presented in Figure 5.2 and range from 60 to 231  $\text{Bq kg}^{-1}$  (d.w.), where the highest activity concentration (317  $\text{Bq kg}^{-1}$  d.w.) was found in one of the monthly samples collected at Utsira. Compared with the results from 1999-2001 (Rudjord *et al.*, 2001; Gäfvert *et al.*, 2003), the levels of  $^{99}\text{Tc}$  have decreased at most sampling sites. The trend can also be seen in Figures 5.3 and 5.4 which show the annual average activity concentration of  $^{99}\text{Tc}$  in *Fucus vesiculosus* at Utsira and Hillesøy, together with the annual discharge of  $^{99}\text{Tc}$  from Sellafield. The maximum levels in Figures 5.3 and 5.4 are the response to the increased discharge from Sellafield in the mid 1990s. From Figures 5.3 and 5.4, one can see that the peak in  $^{99}\text{Tc}$  levels in *F. vesiculosus* occurred after about 3-4 years at Utsira and about 4-5 years at Hillesøy further down-stream. The slight

increase in  $^{99}\text{Tc}$  levels at Utsira in 2004 can be explained by the relatively high discharge of  $^{99}\text{Tc}$  from Sellafield in 2001 and 2002.

At Hillesøy, other seaweed species than *Fucus vesiculosus* were also analysed in 2004 (Table 5.1).

**Table 5.1.** Activity concentration of  $^{99}\text{Tc}$  in seaweed collected at Hillesøy in 2004.

Seaweed species	Activity conc. of $^{99}\text{Tc}$ ( $\text{Bq kg}^{-1}$ d.w.)
<i>Fucus distichus</i>	51
<i>Fucus serratus</i>	105
<i>Ascophyllum nodosum</i>	182

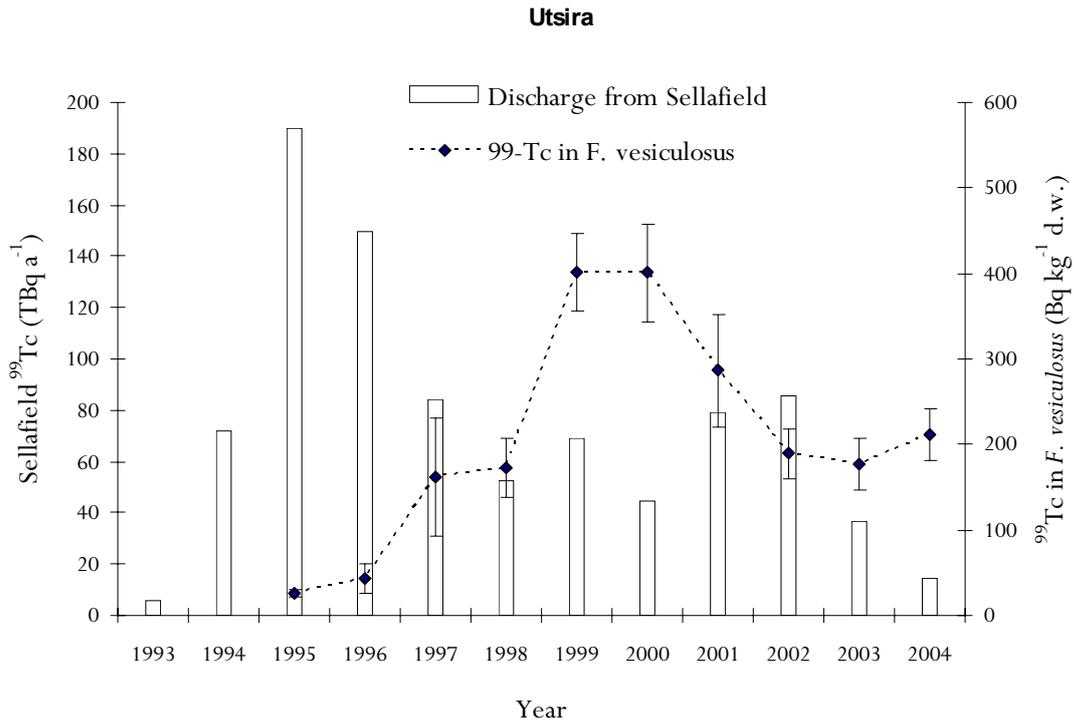


**Figure 5.2.** Levels of  $^{99}\text{Tc}$  in *Fucus vesiculosus* sampled along the Norwegian coastline in 2004.

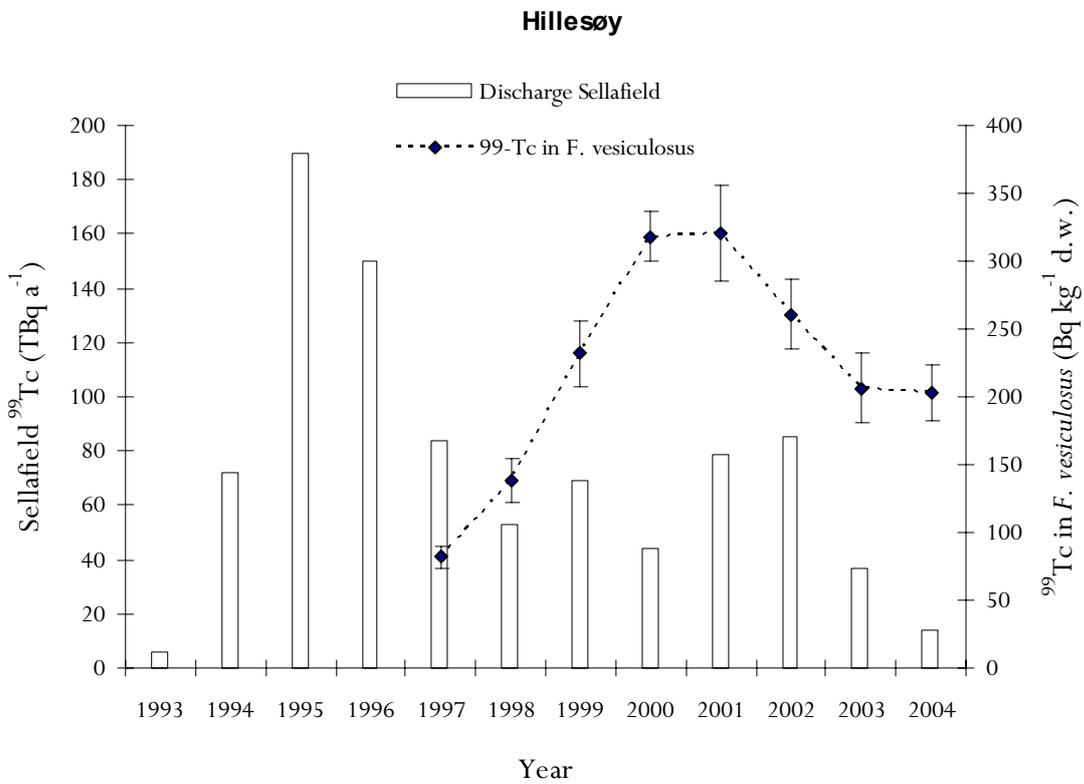
At Hillesøy both seawater and *F. vesiculosus* have been analysed monthly with respect to  $^{99}\text{Tc}$  since 1997. In Figure 5.5 one can see that the activity concentration in the seaweed responded rapidly with the increased  $^{99}\text{Tc}$  levels in the seawater up to mid 2001. From mid 2001 and onwards, a decreasing trend is observed for the

---

<sup>99</sup>Tc level in the water, while the activity concentration in the seaweed has decreased at a slightly slower rate, and shows relatively large fluctuations. Laboratory experiments have shown that the elimination of Tc from macroalgae generally has an initial rapid component (biological  $T_{1/2}$  of 1-3 days) followed by a slower component with a biological  $T_{1/2}$  of 20-200 days (Beasley and Lorz, 1986; Topocuoqlo and Fowler, 1984).



**Figure 5.3.** Annual liquid discharge of  $^{99}\text{Tc}$  from Sellafield (primary axis) and annual average (with 95 % confidence limits)  $^{99}\text{Tc}$  activity concentration in brown algae (*Fucus vesiculosus*) sampled at Utsira (data provided by IFE) in the period 1995-2004 (secondary axis).



**Figure 5.4.** Annual liquid discharge of  $^{99}\text{Tc}$  from Sellafield (primary axis) and annual average (with 95 % confidence limits)  $^{99}\text{Tc}$  activity concentration in brown algae (*Fucus vesiculosus*) sampled at Hillesøy in the period 1997-2004 (secondary axis).

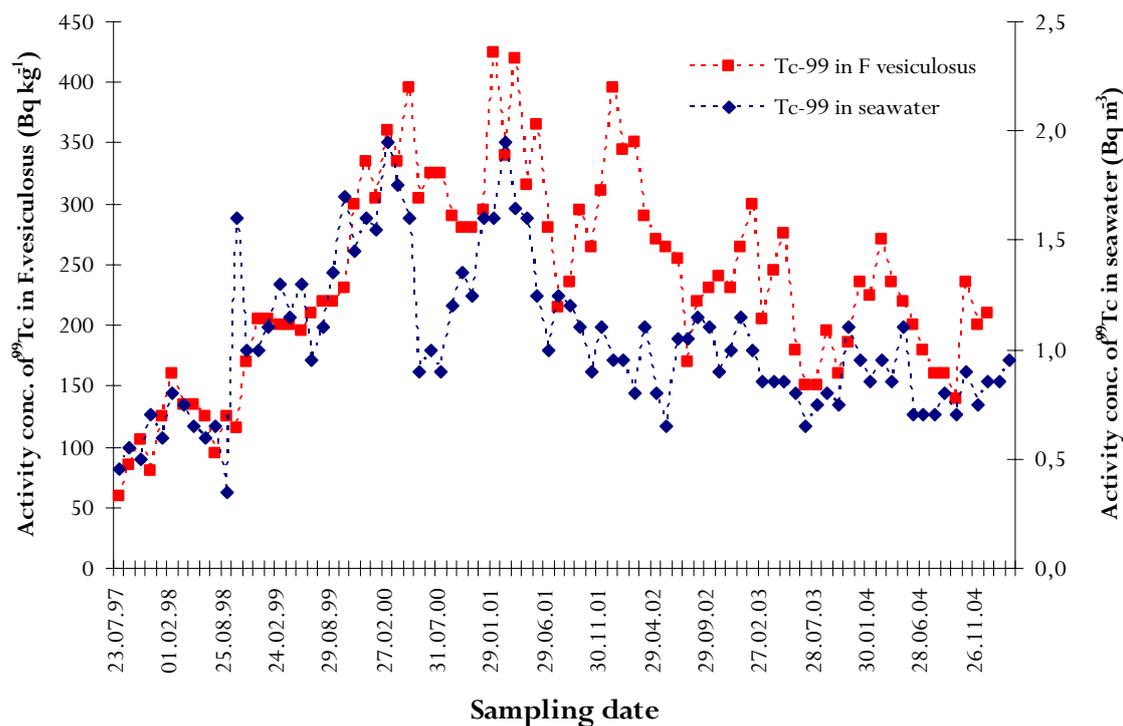
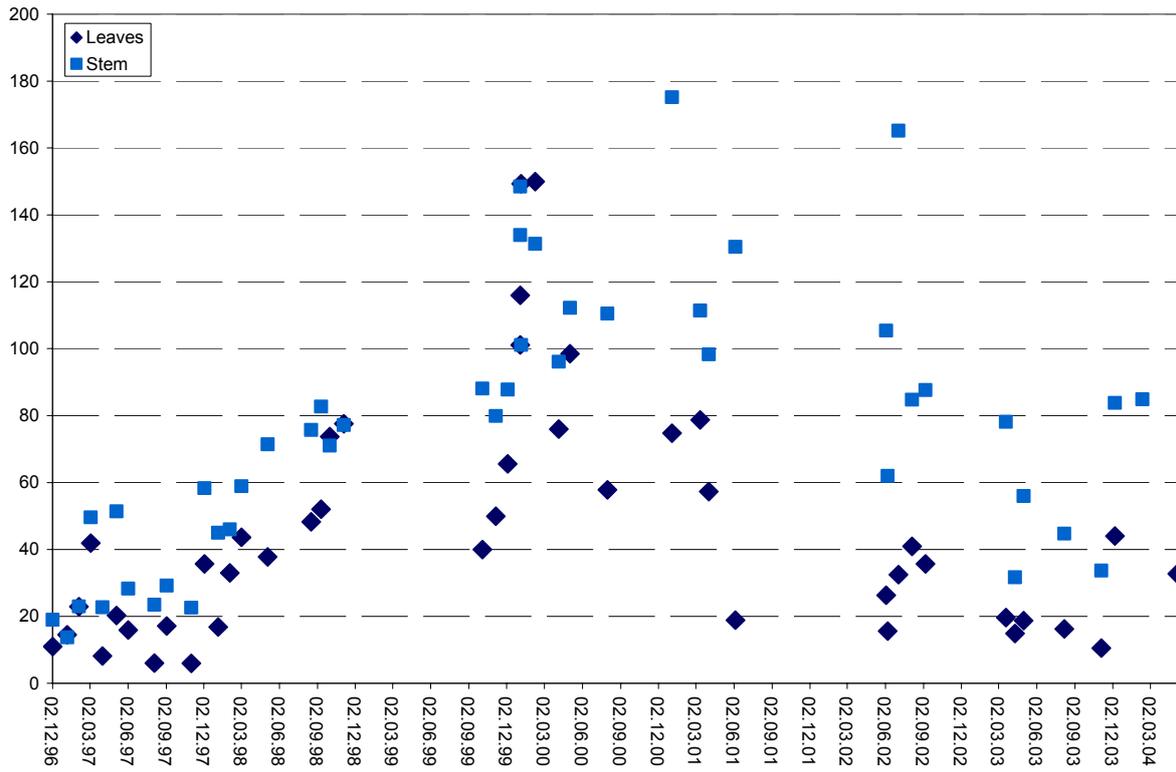


Figure 5.5. Activity concentration of  $^{99}\text{Tc}$  in seawater and seaweed sampled at Hillesøy in the period 1997 to 2004.

Samples of the commercially used algae *Laminaria hyperborea* have been collected and analysed with regard to  $^{99}\text{Tc}$  by the UMB at two different sites on the Norwegian west coast, Kvitsoy and Steinshamn (see Figure 3.1) in the periods 1997 to 2001, and 1999 to 2003, respectively (Mobbs and Salbu, 2002; Jerpetjøn *et al.*, 2005). This alga is routinely harvested along the coastline and used in the alginate industry. *Laminaria hyperborea* is a large brown alga which grows on rocky substrata up to 30 m below the surface. It consists of a stiff, tapering stipe (constituting about 60 % of the biomass), up to 2 m tall, and a broad, deeply divided blade. During the growing season the old blades are shed and replaced with new ones. It can live for up to 15 years. The algae samples were divided into blades and stipes and the results are presented in Figure 5.6. The results show that the activity of  $^{99}\text{Tc}$  increased in both blades and stipes in the period 1997 to early 2000, with a slightly higher CF for the stipes compared with the blades. Since then, the activity has decreased in the blades, which are replaced

annually, while the activity in the stipes has remained at an elevated level. The pattern of activity in the blades suggests that the peak of the discharge plume from Sellafield has reached these sites. However, the higher uptake and slower elimination of  $^{99}\text{Tc}$  in the stipes shows that the levels in the stipes will remain at an elevated level after the plume of  $^{99}\text{Tc}$ -contaminated water has passed these sites. The slight increase in  $^{99}\text{Tc}$  activity concentration in 2004 is likely to be due to the relatively high discharge of  $^{99}\text{Tc}$  from Sellafield in 2001 and 2002.



**Figure 5.6.** Activity concentration ( $Bq\ kg^{-1}\ d.w.$ ) of  $^{99}Tc$  in stipes and blades of *Laminaria hyperborea* sampled at Kvitsoy and Steinshamn (data provided by the UMB).

## 5.2 Caesium-137, strontium-90 and plutonium-239+240 in seaweed

### 5.2.1 Caesium-137 in seaweed

*Fucus vesiculosus* has also been widely used as a bioindicator for  $^{137}\text{Cs}$ . The accumulation of  $^{137}\text{Cs}$  in brown algae is, however, not as pronounced as for  $^{99}\text{Tc}$ . The uptake of  $^{137}\text{Cs}$  also depends on the salinity of the surrounding sea water, with higher uptake at lower salinities (Carlsson and Erlandsson, 1991).

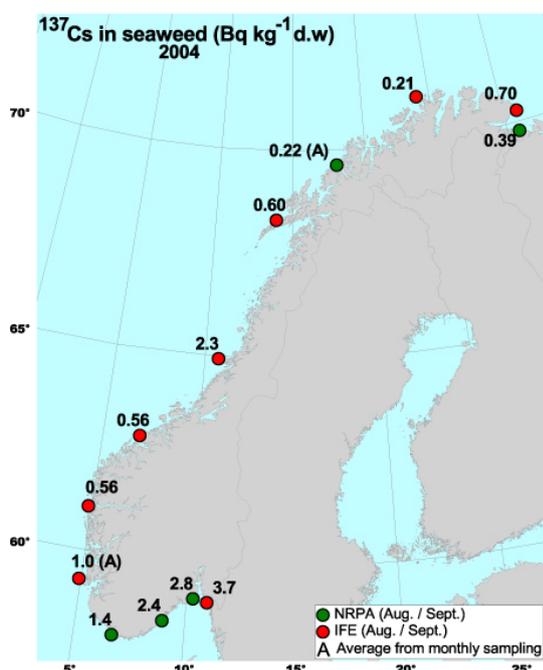
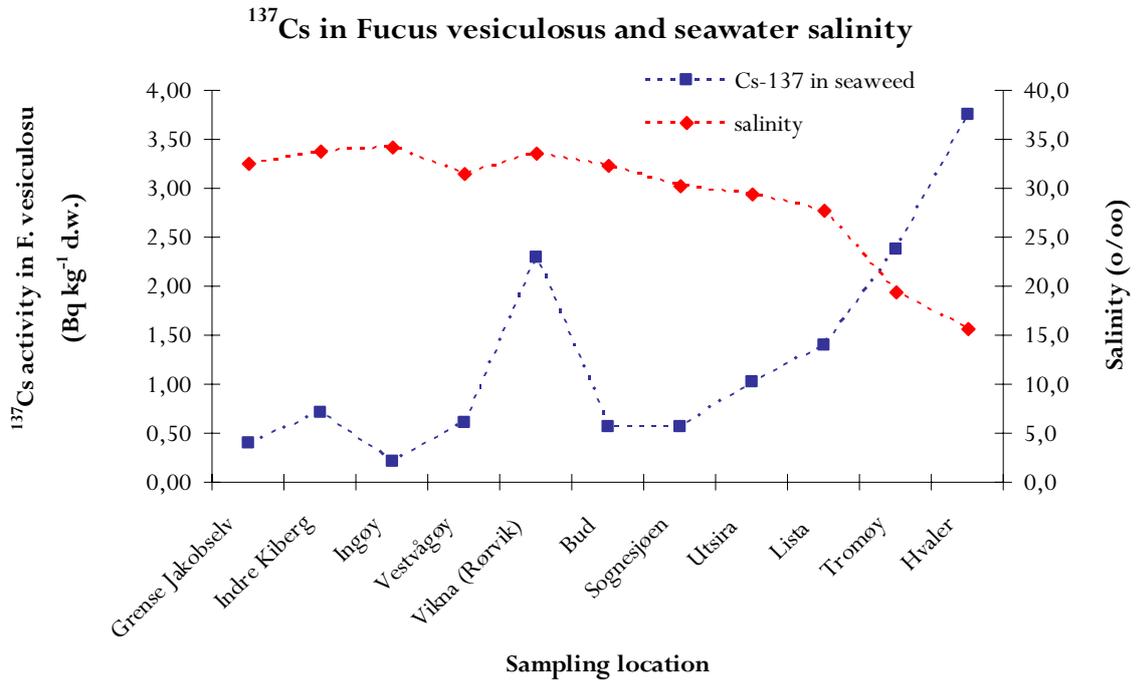


Figure 5.7. Levels of  $^{137}\text{Cs}$  ( $\text{Bq kg}^{-1} \text{ d.w.}$ ) in *Fucus vesiculosus* sampled along the Norwegian coastline in 2004.

In 2004, samples of *Fucus vesiculosus* from the permanent coastal stations (see Figure 3.1) were analysed with respect to  $^{137}\text{Cs}$ . The results are presented in Figure 5.7, and range from 0.21 Bq kg<sup>-1</sup> (d.w.) in the north to 3.7 Bq kg<sup>-1</sup> (d.w.) close to the Swedish border. The reason for the higher activity concentration in *Fucus vesiculosus* in the southern part of Norway is a combination of two effects. First, the salinity in the water is lower and secondly, that the activity concentration of  $^{137}\text{Cs}$  in the seawater is higher due to outflowing Baltic Sea water contaminated by the Chernobyl accident. In Figure 5.8 the activity concentration of  $^{137}\text{Cs}$  in

the seaweed is plotted together with the salinity of the water for 11 of the permanent coastal stations. The peak in the  $^{137}\text{Cs}$  levels in seaweed collected at Vikna can be explained by run-off of  $^{137}\text{Cs}$  from land, since this area was affected by fallout from the Chernobyl accident in 1986.

Comparing those results with the levels found at the coastal sampling stations in 2001, 2002 and 2003 (Gäfvert *et al.*, 2003; NRPA, 2004 and NRPA 2005), one can see that the activity concentration of  $^{137}\text{Cs}$  in *F. vesiculosus* has been relatively stable during recent years. In the southern part of Norway the levels have been in the range 1.2 to 6.0 Bq kg<sup>-1</sup> (d.w.), while in the northern part all results have been below 1 Bq kg<sup>-1</sup> (d.w.).



**Figure 5.8.** Activity concentration ( $Bq\ kg^{-1}\ d.w.$ ) in *Fucus vesiculosus* versus salinity in seawater sampled along the Norwegian coastline in 2004 (data provided by IFE).

---

### 5.2.2 Strontium-90 in seaweed

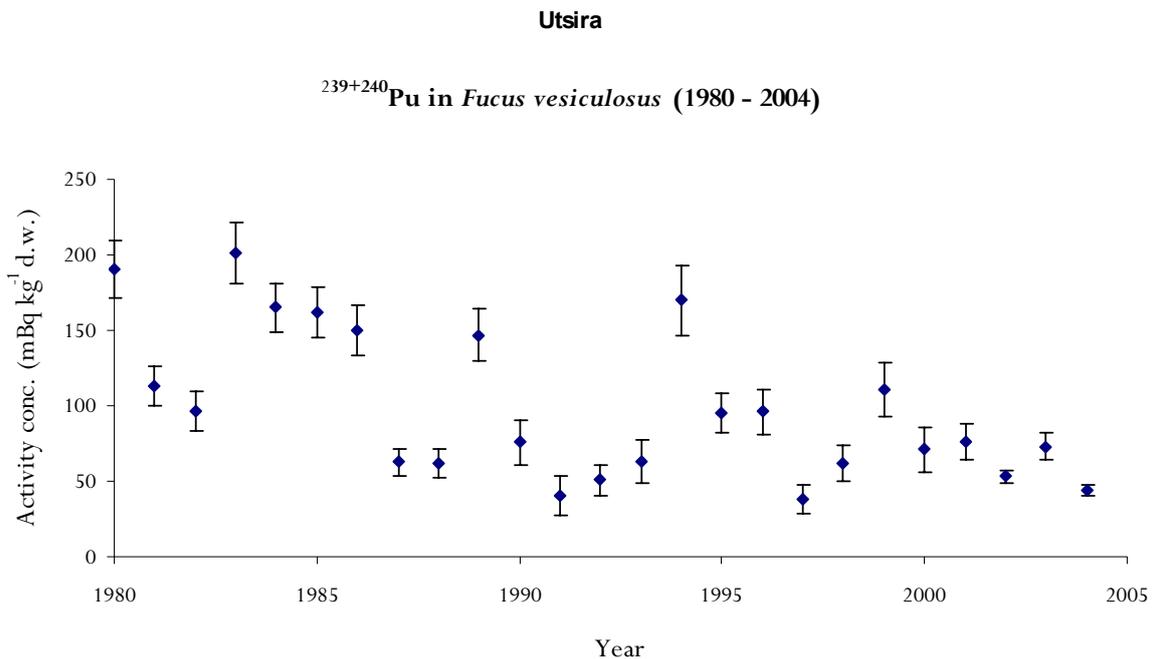
Strontium-90 has been analysed in seaweed samples (*Fucus vesiculosus*) collected at four different sites along the Norwegian coast (Table 5.2). The concentration factor for  $^{90}\text{Sr}$  in seaweed is relatively low. The activity concentration of  $^{90}\text{Sr}$  in seaweed sampled in 2004 ranged from 0.14 to 0.60 Bq kg<sup>-1</sup> (d.w.), where the highest levels were found in seaweed samples from the southern coast of Norway.

**Table 5.2.** Activity concentration of  $^{90}\text{Sr}$  in seaweed (*Fucus vesiculosus*).

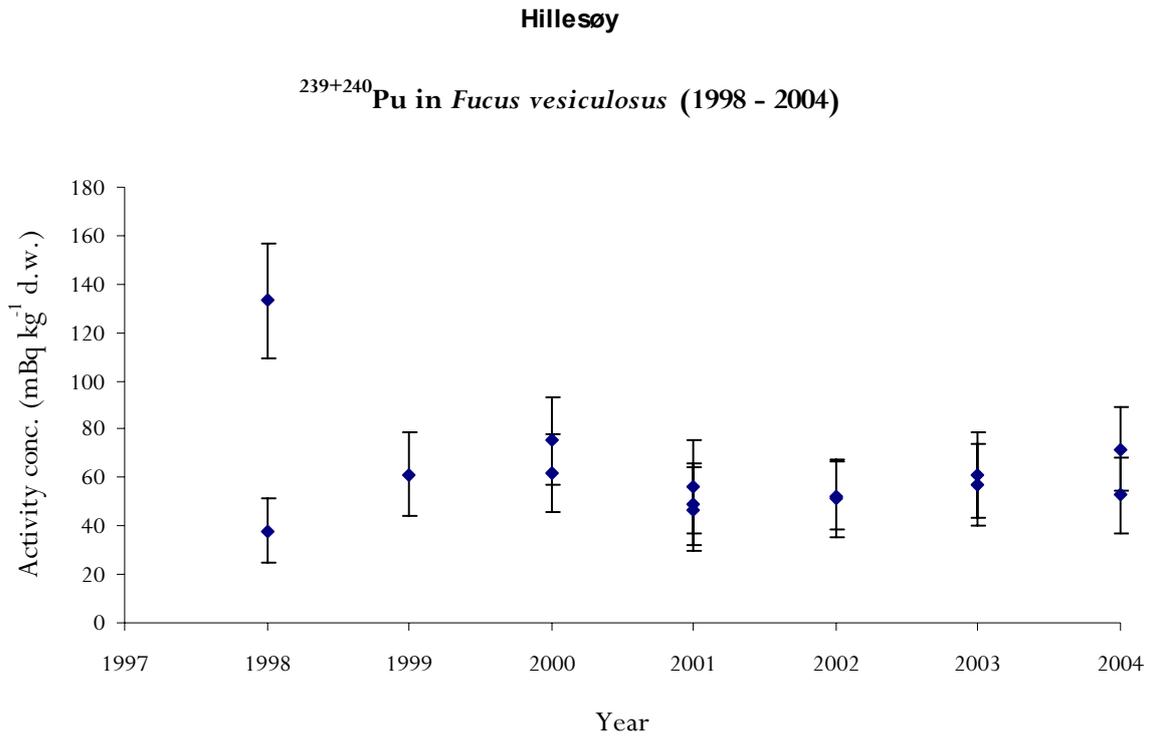
Location	Activity conc. of $^{90}\text{Sr}$ (Bq kg <sup>-1</sup> d.w.)
Lista	0.22
Tjøme	0.60
Hillesøy	0.18
Grense Jacobselv	0.14

### 5.2.3 Plutonium-239+240 in seaweed

*Fucus vesiculosus* has been collected and analysed for  $^{239+240}\text{Pu}$  at Utsira since 1980. The results from the period 1980 to 2004 are presented in Figure 5.9. The activity concentrations in these samples were in the range 40 to 201  $\text{mBq kg}^{-1}$ , with relatively large fluctuations from year to year. Two samples collected at Hillesøy in Mars and August 2004 showed an average activity concentration of 62  $\text{mBq kg}^{-1}$  (d.w.). In Figure 5.10 the activity concentration of  $^{239+240}\text{Pu}$  in *Fucus vesiculosus* at Hillesøy since 1998 is presented. One can also see that the level of  $^{239+240}\text{Pu}$  has remained at the same level in recent years. The levels are similar to those found by Roos *et al.* (1993) in the late 1980s and early 1990s off the Swedish west coast. At this site (Särdal) levels up to around 600  $\text{mBq kg}^{-1}$  (d.w.) were found in *Fucus vesiculosus* in the 1970s due to the higher discharge of plutonium from Sellafield during the 1960s and 1970s.



**Figure 5.9.**  $^{239+240}\text{Pu}$  levels ( $\text{mBq kg}^{-1}$  d.w.) in *Fucus vesiculosus* at Utsira in the period 1980 to 2004 (data provided by IFE).



**Figure 5.10.**  $^{239+240}\text{Pu}$  levels ( $\text{mBq kg}^{-1}$  d.w.) in *Fucus vesiculosus* at Hillesøy in the period 1998 to 2004.

### 5.3 Technetium-99 and caesium-137 in crustaceans and molluscs

In 2004, samples of lobster (*Homarus gammarus*) were collected at the coastal station in Arendal (see Figure 5.11) and analysed for  $^{99}\text{Tc}$ . Lobsters have previously been identified as a marine organism with a high ability to accumulate  $^{99}\text{Tc}$  from seawater (Swift, 1985; Smith *et al.*, 1998; Smith *et al.*, 2001). The observed levels are presented in Table 5.3 and range from 2.0 to 70.0 Bq kg<sup>-1</sup> w.w. The levels are similar to those found in 2001, 2002 and 2003 in lobster samples from Rogaland (Kolstad and Lind, 2002) and Tisler, Arendal and Vaerlandet in 2002 and 2003 (NRPA, 2004; NRPA, 2005b). The results show, however, a relatively large variation, even for samples collected in the same area. The activity concentration of  $^{99}\text{Tc}$  in the seawater in the different sampling areas is in the range 1.2 to 1.6 Bq m<sup>-3</sup>. Furthermore, it is observed a clear difference between female and male lobsters where female lobsters have a significantly higher uptake of  $^{99}\text{Tc}$ .



Figure 5.11. Geographic overview of the sampling areas.

Table 5.3. Average activity concentration and range of  $^{99}\text{Tc}$  in lobster (*Homarus gammarus*) tail muscle in 2004.

Location	Gender (F/M)	n	$^{99}\text{Tc}$ Average (range) (Bq kg <sup>-1</sup> w.w.)
Arendal	F	3	38.2 (19.5 - 70.0)
	M	4	3.9 (2.0 - 5.0)

Some lobster samples were also analysed for  $^{137}\text{Cs}$ . The levels were considerably lower than for  $^{99}\text{Tc}$  and ranged from < 0.1 to 0.32 Bq kg<sup>-1</sup> (w.w.).

Mussels (*Mytilus edulis*) were collected in 2004 in coastal regions from the southern coast of Norway (see Figure 5.11). From each region about 0.5 kg (w.w.) of the mussel soft tissue was collected, homogenized and later analysed for  $^{137}\text{Cs}$ . The results are presented in Table 5.4.

Table 5.4. Activity concentration of  $^{137}\text{Cs}$  in Blue mussels and Common limpet in 2004.

Location/Species	$^{137}\text{Cs}$ (Bq kg <sup>-1</sup> w.w.)
Vestfold/Blue mussels ( <i>Mytilus edulis</i> )	0.10 ± 0.02
Vest-Agder/Common Limpet ( <i>Patella vulgata</i> )	0.08 ± 0.02

Samples of Kamtchatca crab (*Paralithodes camtschaticus*) and shrimps (*Pandalus borealis*) from Norwegian and adjacent marine waters have been collected and analysed for  $^{137}\text{Cs}$ . The results and the sampling location are presented in Table 5.5.

**Table 5.5.** Activity concentration ( $Bq\ kg^{-1}\ w.w.$ ) in various marine species 2004.

Species	Location	$^{137}Cs$ ( $Bq\ kg^{-1}\ w.w.$ )
Shrimps ( <i>Pandalus borealis</i> )	Fleinvær, Nordland	< 0.10
Shrimps ( <i>Pandalus borealis</i> )	North Sea	$0.20 \pm 0.01$ to $0.01 \pm 0.10$
Kamtchatca crab ( <i>Paralithodes camtschaticus</i> )	Varangerfjorden Finnmark	< 0.10
Sea Cucumbers ( <i>Holothuroidea</i> )	North Sea	$0.20 \pm 0.01$

## 5.4 Caesium-137 in fish

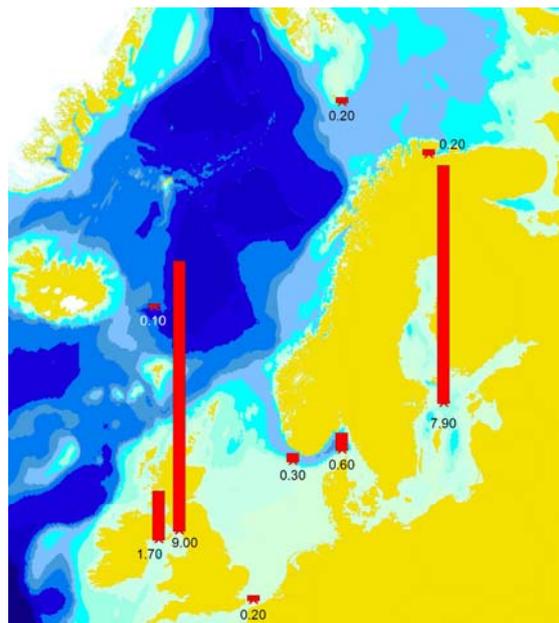
Commercially important fish species, including farmed salmon, as well as other fish species have been collected from Norwegian marine waters and analysed with respect to  $^{137}Cs$ .

The activity concentrations of  $^{137}Cs$  in fish caught in the Barents Sea, the Norwegian Sea, the North Sea and in Skagerrak are presented in Table 5.7. The levels of  $^{137}Cs$  in all sampled fish species from these areas range from 0.1 to 0.8  $Bq\ kg^{-1}$  (w.w.).

The activity concentrations of  $^{137}Cs$  in various fish species collected along the Norwegian coastline from Finnmark in the north to the Oslofjord in the south are given in Table 5.8. The results from this area range from 0.1 to 1.7  $Bq\ kg^{-1}$  (w.w.).

Generally, the levels of  $^{137}Cs$  in Norwegian marine waters can be considered low. In the marine environment surrounding Norway the highest concentrations of  $^{137}Cs$  are found in fish caught next to the Norwegian coastline. These results could be explained as a result of Chernobyl run-off of  $^{137}Cs$  from land and outflow of Chernobyl fallout from the Baltic Sea to the Norwegian coastal Current.

In Figure 5.12, typical activity concentrations of  $^{137}Cs$  found in cod in northern European waters are presented.



**Figure 5.12.** Levels of  $^{137}Cs$  ( $Bq\ kg^{-1}\ w.w.$ ) in cod from different marine areas (data from EA, EHS, FSA & SEPA (2004) and this report).

In the Baltic Sea, the levels of  $^{137}Cs$  in fish are higher due to higher levels of  $^{137}Cs$  in the water, but also due to the lower salinity of the water, resulting in a higher uptake in fish. In the Irish Sea, the levels are influenced by past and present discharges of  $^{137}Cs$  from the reprocessing plant at Sellafield.

### 5.4.1 Farmed fish

In Table 5.6 the results from the analyses of  $^{137}Cs$  in farmed salmon (*Salmo salar*) from locations along the northern Norwegian coast are presented. The activity concentration of  $^{137}Cs$  ranges from < 0.1 to  $0.3 \pm 0.1\ Bq\ kg^{-1}$  (w.w.).

**Table 5.6.** Levels of  $^{137}Cs$  in farmed salmon in 2004.

Location	$^{137}Cs$ in muscle tissue ( $Bq\ kg^{-1}\ w.w.$ )
Finnmark	$0.26 \pm 0.09$
Troms	$0.24 \pm 0.11$
Nordland	< 0.10

**Table 5.7.** Levels of  $^{137}\text{Cs}$  activity concentrations ( $\text{Bq kg}^{-1}$  w. w.) for various fish species collected from the Barents Sea, Norwegian Sea, the North Sea and in the Skagerrak, in 2004.

Species	Location	No. of samples (total no. of fish)	$^{137}\text{Cs}$ in muscle tissue ( $\text{Bq kg}^{-1}$ w.w.)
Cod ( <i>Gadus morhua L.</i> )	Barents Sea	3 (75)	$0.20 \pm 0.01$ to $0.4 \pm 0.10$
Saithe ( <i>Pollachius virens</i> )		2 (50)	$0.20 \pm 0.01$ and $0.23 \pm 0.07$
Long Rough Dab ( <i>Hippoglossoides Platessoides</i> )		1(5)	<0.4
Golden Redfish ( <i>Sebastes Marinus</i> )		1(5)	<0.3
Deep-Sea Redfish ( <i>Sebastes mentella</i> )		1(5)	$0.20 \pm 0.01$
Greenland Halibut ( <i>Reinhardtius Hippoglossoides</i> )		1(5)	$0.30 \pm 0.01$
Atlantic herring ( <i>Clupea harengus</i> )	Norwegian Sea	2 (10)	$0.14 \pm 0.04$ and $0.33 \pm 0.07$
Mackerel ( <i>Scomber scombrus</i> )		2 (10)	$0.24 \pm 0.06$ and $0.30 \pm 0.07$
Cod ( <i>Gadus morhua L.</i> )	The North Sea	3 (15)	$0.11 \pm 0.02$ to $0.30 \pm 0.10$
Saithe ( <i>Pollachius virens</i> )		2 (8)	$0.40 \pm 0.10$ and $0.50 \pm 0.10$
Atlantic herring ( <i>Clupea harengus</i> )		11 (234)	$0.20 \pm 0.08$ to $0.47 \pm 0.11$
Haddock ( <i>Melanogrammus Aeglefinus.</i> )		1 (2)	$0.30 \pm 0.01$
Mackerel ( <i>Scomber scombrus</i> )		15 (92)	$0.12 \pm 0.04$ to $0.39 \pm 0.08$
Long Rough Dab ( <i>Hippoglossoides Platessoides</i> )		1 (18)	$0.10 \pm 0.01$
Hake ( <i>Merluccius Merluccius</i> )		1 (5)	$0.60 \pm 0.01$
Blue Whiting ( <i>Micromesistius Poutassou</i> )		4 (66)	$0.30 \pm 0.10$ to $0.4$
Norway pout ( <i>Trisopterus Esmarkii</i> )		2	$0.10 \pm 0.10$ and $0.20 \pm 0.10$
Vahl's eelpout ( <i>Lycodes Vahli</i> )		1	$0.10 \pm 0.01$
Cod ( <i>Gadus morhua L.</i> )		Skagerrak	4 (17)
Saithe ( <i>Pollachius virens</i> )	4 (19)		$0.40 \pm 0.10$ to $0.60 \pm 0.10$
Haddock ( <i>Melanogrammus Aeglefinus.</i> )	1 (2)		$0.20 \pm 0.01$
European plaice ( <i>Pleuronectes Platessa</i> )	1 (17)		< 0.5
Blue Whiting ( <i>Micromesistius Poutassou</i> )	3 (48)		$0.30 \pm 0.10$ to $0.40 \pm 0.10$
Norway pout ( <i>Trisopterus Esmarkii</i> )	3		< 0.2
Vahl's eelpout ( <i>Lycodes Vahli</i> )	2		$0.10 \pm 0.10$ and $0.20 \pm 0.10$

**Table 5.8.** Levels of  $^{137}\text{Cs}$  activity concentrations ( $\text{Bq kg}^{-1}$  w. w.) for various fish species collected off the coast of Finnmark in the north to the Oslo fjord in the south.

Species	Location	No. of samples (total no. of fish)	$^{137}\text{Cs}$ in muscle tissue ( $\text{Bq kg}^{-1}$ w.w.)
Cod ( <i>Gadus morhua</i> L.)	Coastal waters of Finnmark and Troms	3 (32)	$0.30 \pm 0.01$
Saithe ( <i>Pollachius virens</i> )		3 (35)	$0.25 \pm 0.09$ to $0.30 \pm 0.10$
Haddock ( <i>Melanogrammus Aeglefinus.</i> )		3 (15)	$< 0.14$ to $0.30 \pm 0.10$
Saithe ( <i>Pollachius virens</i> )	Coastal waters of Nordland and Trøndelag	8(180)	$< 0.1$ to $0.46 \pm 0.17$
Haddock ( <i>Melanogrammus Aeglefinus.</i> )		1 (25)	$0.10 \pm 0.10$
Cod ( <i>Gadus morhua</i> L.)	Coastal waters of the eastern Norway	3 (10)	$0.80 \pm 0.10$ to $1.70 \pm 0.10$
Haddock ( <i>Melanogrammus Aeglefinus.</i> )		1 (5)	$0.30 \pm 0.01$
Atlantic herring ( <i>Clupea harengus</i> )		4 ( 20)	$0.60 \pm 0.10$ to $0.80 \pm 0.10$
Sprat ( <i>Sprattus Sprattus</i> )		3	$0.70 \pm 0.10$ to $0.90 \pm 0.10$
Whiting ( <i>Merlangius Merlangus</i> )		4 (20)	$0.60 \pm 0.10$ to $1.20 \pm 0.10$
European Anchovy ( <i>Engraulis Encrasicolus</i> )		1	$<0.4$
Hake ( <i>Merluccius Merluccius</i> )		1 (5)	$0.90 \pm 0.10$

## 6 Summary and conclusions

In 2004 samples of seawater, sediment, and seaweed were collected in the North Sea, the Skagerrak, around Svalbard, and at a number of coastal stations, including those off the islands Bjørnøya, Hopen and Jan Mayen. Data on radioactivity levels and trends in these areas together with data on liquid discharges from nuclear installations and recent trends in such discharges are summarised in this report.

### 6.1 Sources

The liquid discharges from the nuclear facilities of IFE at Kjeller and in Halden in 2004 are within authorised limits according to the estimates made by the operator. The effective dose to a member of the critical group from liquid radioactive discharge at Kjeller was estimated to be 0.07  $\mu\text{Sv}$  in 2004, which correspond to 7 % of the dose limit. The effective dose to the critical group from IFE Halden liquid radioactive discharge was estimated to be 0.016  $\mu\text{Sv}$  in 2004, corresponding to 1.6 % of the dose limit. The discharge of anthropogenic radionuclides from Norwegian sources is only detectable in the environment close to each discharge point and has no significant impact on the large-scale distribution of these radionuclides in the marine environment.

Unsealed radioactive substances are used in hospitals, research laboratories and various industrial activities. Among the most abundant radionuclides are  $^{99\text{m}}\text{Tc}$ ,  $^{131}\text{I}$  and  $^3\text{H}$ . Regarding the radiological impact on the public of the discharge of these substances,  $^{131}\text{I}$  is one of the most important radionuclides. The amount of  $^{131}\text{I}$  sold in Norway in 2004 was 1.889 TBq.

Produced water from offshore oil production may contain enhanced levels of naturally occurring radium isotopes. The reported total discharge of  $^{226}\text{Ra}$  in produced water from Norwegian platforms in 2004 was 445 GBq,

where about 45 % was discharged from 2 platforms, Troll B and Troll C.

The long-range transports of radionuclides originating from nuclear weapons fallout, the Chernobyl accident and from reprocessing of spent nuclear fuel are still the main contributors to anthropogenic radionuclides found in Norwegian waters. In addition to direct discharges from reprocessing, water from the Baltic Sea and remobilisation of  $^{137}\text{Cs}$  and plutonium from contaminated Irish Sea sediments act as secondary sources of radionuclides to the Norwegian marine environment.

### 6.2 Radioactivity in seawater and sediment

In 2004, samples of seawater and sediment were collected in the North Sea and the Skagerrak and analysed with respect to  $^{137}\text{Cs}$ ,  $^{99}\text{Tc}$ ,  $^{90}\text{Sr}$ ,  $^{238}\text{Pu}$ ,  $^{239+240}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ . A general trend seen in most samples is that the levels of radioactivity are similar, or slightly lower than have been observed in recent years. One exception is  $^{137}\text{Cs}$  where significantly higher levels were found in the Skagerrak in 2004 compared with levels observed in 2001. For the first time a large survey of radium in the North Sea and the Skagerrak has also been included.

#### 6.2.1 Caesium-137 in seawater and sediment

Observed levels of  $^{137}\text{Cs}$  in surface water in the North Sea, the Skagerrak and along the Norwegian coast in 2004 ranged from 1.6 to 29.6  $\text{Bq m}^{-3}$ . The highest activity concentrations were found in the Skagerrak in May. The correlation between activity concentration and salinity in the seawater shows that the higher activity concentration is due to outflowing seawater from the Baltic Sea. The activity concentration of  $^{137}\text{Cs}$  in sediment from the North Sea and the Skagerrak ranged from  $< 0.4$  to 13.4  $\text{Bq kg}^{-1}$  (d.w.), which is similar to what was found in 2001 in the same area.

### 6.2.2 Technetium-99 in seawater

Samples of seawater were collected in the North Sea, the Skagerrak and around Svalbard, Jan Mayen, Hopen and Bjørnøya. Levels of  $^{99}\text{Tc}$  in surface water ranged from 0.11 to 3.3 Bq m<sup>-3</sup> in the northern North Sea and the Skagerrak and from 0.06 to 0.22 Bq m<sup>-3</sup> in seawater sampled around Svalbard, Jan Mayen Hopen and Bjørnøya. The highest concentrations were found off the coast of Scotland, as expected. The levels in the North Sea and the Skagerrak are similar to those found in 2001. The levels around Svalbard in 2004 are generally lower than those reported for this region in 2000 and 2001. The annual average activity concentration obtained from monthly sampling at Hillesøy shows a similar activity concentration in 2004 as in 2003.

### 6.2.3 Strontium-90 in seawater

Seawater samples collected in the North Sea and the Skagerrak showed activity concentrations in the range 1.0 to 6.1 Bq m<sup>-3</sup>. The highest levels in 2004 were found in samples from the Skagerrak. The slightly higher levels in 2004 compared to 2001, are most likely caused by outflowing seawater from the Baltic Sea.

### 6.2.4 Plutonium-239+240 and americium-241 in seawater

Observed levels of  $^{239+240}\text{Pu}$  in the North Sea and the Skagerrak in 2004 ranged from 3.5 to 25.6 mBq m<sup>-3</sup>, where the highest level was found off the coast of Scotland. The activity concentrations of  $^{241}\text{Am}$  in seawater samples from the same area were generally lower and ranged from 1.1 to 20.5 mBq m<sup>-3</sup>. The observed levels of Pu in the North Sea in 2004 were lower than those found in the same area in 2001.

### 6.2.5 Radium-226 and radium-228 in seawater

Radium-226 and radium-228 were analysed in seawater samples collected in the northern North Sea and in the Skagerrak. The activity concentration of  $^{226}\text{Ra}$  in the samples ranged from 1.0 to 2.9 Bq m<sup>-3</sup>, where the highest levels were found in coastal water and in the

Skagerrak. The  $^{228}\text{Ra}/^{226}\text{Ra}$  ratio ranged from 0.1 in Atlantic water to 2.0 in coastal water.

## 6.3 Radioactivity in biota

### 6.3.1 Technetium-99 in seaweed

Samples of *Fucus vesiculosus* sampled at the permanent coastal stations showed activity concentrations in the range 60 to 231 Bq kg<sup>-1</sup> (d.w.). For most stations the levels were similar in 2004 compared to 2002 and 2003. Monthly sampling at Hillesøy, Utsira, and Steinshamn all indicate that the peak of  $^{99}\text{Tc}$  from the Sellafield discharge in 1995 and 1996 has passed these sites. The activity concentration of  $^{99}\text{Tc}$  in seaweed from Utsira showed a slight increase in 2004 compared to 2003, which can be explained by the relatively high Sellafield discharge of  $^{99}\text{Tc}$  in 2001 and 2002.

### 6.3.2 Caesium-137 in seaweed

The activity concentration of  $^{137}\text{Cs}$  in *F. vesiculosus* sampled at the coastal stations was in the range 0.21 to 3.7 Bq kg<sup>-1</sup> (d.w.), where the highest levels were found in the southern part of Norway, where the impact from outflowing Baltic Sea water is highest. Apart from a higher activity concentration of  $^{127}\text{Cs}$  in the water, lower salinity also leads to a higher uptake of  $^{137}\text{Cs}$  in the seaweed. A comparison with results from 2000 and 2001 shows that the levels have been relatively stable in recent years.

### 6.3.3 Plutonium-239+240 in seaweed

At two sites, Hillesøy and Utsira, seaweed (*F. vesiculosus*) has also been analysed for  $^{239+240}\text{Pu}$ . The activity concentration of  $^{239+240}\text{Pu}$  in seaweed at these locations has in the period 1998 - 2004 been in the range from 40 to 130 mBq kg<sup>-1</sup> (d.w.). Annual sampling at Utsira since 1980 reveals relatively large fluctuations of  $^{239+240}\text{Pu}$  in seaweed samples.

### 6.3.4 Technetium-99 and caesium-137 in crustaceans and molluscs

Samples of lobster (*Hommarus gammarus*), mussels (*Mytilus edulis*) and other marine species have been sampled at different locations and analysed for  $^{137}\text{Cs}$  and  $^{99}\text{Tc}$ . The activity

---

concentrations of  $^{137}\text{Cs}$  in all analysed samples were low,  $< 0.4 \text{ Bq kg}^{-1}$  (w.w.). Technetium-99 was analysed in lobsters where the average level in female lobsters was  $38 \text{ Bq kg}^{-1}$  (w.w.) and  $4 \text{ Bq kg}^{-1}$  in male lobsters.

#### ***6.3.5 Caesium-137 in fish***

Different species of fish, commercially important and others, have been sampled in the Barents Sea, Norwegian Sea, the North Sea and the Skagerrak. The activity concentrations of  $^{137}\text{Cs}$  in fish are generally low, below  $1 \text{ Bq kg}^{-1}$  (w.w.). The highest measured activity concentration was found in cod sampled in coastal waters in the eastern part of Norway.

## References

- Amundsen et al., 2003  
Amundsen I, Brekken A, Liland A. Utslipp av radioaktive stoffer fra Sellafield-anleggene. En gjennomgang av britiske myndigheters regulering av utslippstillatelser. StrålevernRapport 2003:2. Østerås: Statens strålevern, 2003 (In Norwegian, English summary).  
[http://www.nrpa.no/dokumentarkiv/Straleve\\_rnrapport2\\_2003.pdf](http://www.nrpa.no/dokumentarkiv/Straleve_rnrapport2_2003.pdf) (06.01.06)
- Aure et al., 1998  
Aure J, ed. Havets miljø 1998. Fisken og havet, særnr. 2-1998. Bergen: Havforskningsinstituttet, 1998. (In Norwegian)
- Beasley and Lorz, 1986  
Beasley TM, Lorz HV. A review of the biological and geochemical behaviour of technetium in the marine environment. *Journal of Environmental Radioactivity* 1986; 3: 1-22.
- Bourlat et al., 1996  
Bourlat Y, Milliès-Lacroix J-C, Le Petit G, Bourguignon J.  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and  $^{239+240}\text{Pu}$  in world ocean water samples collected from 1992 to 1994. In: Guéguéniat P, Germain P, Métivier H, eds. Radionuclides in the oceans. Les Ulis: Institute de Protection et de Sureté Nucleaire / Les editions de physique, 1996: 75-93.
- Brown et al., 2002  
Brown J, Iospje M, Kolstad AK., Lind B, Rudjord AL, Strand P. Temporal trends for  $^{99}\text{Tc}$  in Norwegian coastal environments and spatial distribution in the Barents Sea. *Journal of Environmental Radioactivity* 2002; 60: 49-60.
- Carlsson L. and Erlandsson B., 1991  
Carlsson L, Erlandsson B. Effects of salinity on the uptake of radionuclides by *Fucus vesiculosus* L. *Journal of Environmental Radioactivity* 1991; 13: 309-322.
- Chen et al., 1991  
Chen Q et al. Determination of plutonium in environmental samples by controlled valence in anion exchange. RISØ-M-2856. Roskilde: Risø National Laboratory, 1991.
- Chen et al., 2001  
Chen Q, Aakrog A, Nielsen SP, Dahlgaard H, Lind B, Kolstad AK, Yu, Y. Procedures for determination of  $^{239,240}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{237}\text{Np}$ ,  $^{234,238}\text{U}$ ,  $^{228,230,232}\text{Th}$ ,  $^{99}\text{Tc}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  in environmental materials. Risø-R-1263. Roskilde: Risø National Laboratory, 2001.
- Cook et al., 1997  
Cook GT, Mac Kenzie AB, Mc Donald P and Jones SR. Remobilization of Sellafield-derived radionuclides and transport from the North-East Irish Sea. *Journal of Environmental Radioactivity* 1997; 35: 227-241.
- Dahlgaard, 1995  
Dahlgaard. (1995). Transfer of European Coastal Pollution to the Arctic: Radioactive tracers. *Marine Pollution Bulletin*, Vol. 31, Nos 1-3, pp 3-7.
- Dahlgaard, 2002  
Dahlgaard H. Baltic  $^{137}\text{Cs}$  outflow through the Danish Straits indicates remobilisation. In: Strand P, Børretzen P, Jølle T, eds. International conference on radioactivity in the environment, Monaco 2002. Extended abstracts. Østerås: Norwegian Radiation Protection Authority, 2002: 516-520.
- EA, EHS, FSA & SEPA, 2005  
EA, EHS, FSA & SEPA (Environment Agency, Environment and Heritage Service, Food Standards Agency & Scottish Environment Protection Agency. Radioactivity in food and the environment 2004. RIFE-10. Preston 2005.
- Evans, 1991  
Evans S. Impacts of the Chernobyl fallout in the Baltic Sea ecosystem. In: Moberg L, ed. The Chernobyl fallout in Sweden. Stockholm: Swedish Radiation Protection Institute, 1991: 109-127.
- Gwynn et al., 2004.  
Gwynn JP, Dowdall M, Lind B. The radiological environment of Svalbard.

StrålevernRapport 2004:2. Østerås: Norwegian Radiation Protection Authority, 2004.

Gäfvert et al. 2003

Gäfvert T, Føyn L, Brungot AL, Kolstad AK, Lind B, Christensen GC, Strålberg E, Drefvelin J, Rudjord AL.. Radioactivity in the marine environment 2000 and 2001. Results from the Norwegian National Monitoring Programme (RAME). StrålevernRapport 2003:8. Østerås: Norwegian Radiation Protection Authority, 2003.

[http://www.nrpa.no/dokumentarkiv/StralevernRapport8\\_2003.pdf](http://www.nrpa.no/dokumentarkiv/StralevernRapport8_2003.pdf) (06.01.06)

IAEA, 1990

IAEA (International Atomic Energy Agency). The Environmental Behaviour of Radium. Vol. 1. Technical Reports Series No. 310. ISBN 92-0-125090-9. Vienna, 1990

IFE, 2001a

IFE. Måling av radioaktive stoffer i utslipp til luft, i avløpsvann og i prøveobjekter fra IFE Kjellers omegn og Nitelva. Rapport for 2000. IFE Rapport nr: IFE/I-2001/005. Kjeller: Institutt for energiteknikk, IFE, 2001.

IFE, 2002a

IFE. Måling av radioaktive stoffer i utslipp til luft, i avløpsvann og i prøveobjekter fra IFE Kjellers omegn og Nitelva. Rapport for 2001. IFE Rapport nr: IFE/I-2002-09. Kjeller: Institutt for energiteknikk, IFE, 2002.

IFE, 2003a

IFE. Kontroll av radioaktive stoffer i utslipp til luft og vann og i prøveobjekter fra IFE Kjellers omegn, Nitelva og Himdalen. Rapport for 2002. IFE Rapport nr: IFE/I-2003/016. Kjeller: Institutt for energiteknikk, IFE, 2003.

IFE, 2004a

IFE. Kontroll av radionuklider i utslipp til luft og vann og i prøveobjekter fra IFE Kjellers omegn, Nitelva og Himdalen. Rapport for 2003. IFE Rapport nr: IFE/I-2004/012. Kjeller: Institutt for energiteknikk, IFE, 2004.

IFE, 2005a

IFE. Kontroll av radionuklider i utslipp til luft og vann og i prøveobjekter fra IFE Kjellers omegn, Nitelva og Himdalen. Rapport for 2004. IFE Rapport nr: IFE/I-2005/009. Kjeller: Institutt for energiteknikk, IFE, 2005.

IFE, 2001b

IFE. Utslipp fra Haldenreaktoren i 2000. Sv-rapport 458. Halden: Institutt for energiteknikk, IFE, 2001.

IFE, 2002b

IFE. Utslipp fra Haldenreaktoren i 2001. Sv-rapport 511. Halden: Institutt for energiteknikk, IFE, 2002.

IFE, 2003b

IFE. Utslipp fra Haldenreaktoren i 2002. Sv-rapport 541. Halden: Institutt for energiteknikk, IFE, 2003.

IFE, 2004b

IFE . Utslipp fra Haldenreaktoren i 2003. Sv-rapport 586. Halden: Institutt for energiteknikk, IFE, 2004.

IFE, 2005b

IFE. Utslipp fra Haldenreaktoren I 2004. Sv-rapport 629. Halden: Institutt for energiteknikk, IFE, 2005.

Jerpetjøn et al., 2005

Jerpetjøn A, Salbu B, Oughton D, Skipperud L. (2005). Seaweed, fish and crustaceans as bioindicators for <sup>99</sup>Tc released to the marine environment. To be published in Proceedings of NKS-B Seminar held in Tartu, Estonia, 24-25 October 2005.

Kershaw et al., 1999.

Kershaw PJ, McCubbin D, Leonard KS. Continuing contamination of North Atlantic and Arctic waters by Sellafield radionuclides. The Science of the Total Environment 237-238: 119-132.

Kolstad, 1995

Kolstad AK. Expeditions to «Komsomolets» in 1993 and 1994. StrålevernRapport 1995:7.

Østerås: Norwegian Radiation Protection Authority. (In Norwegian)

Leonard et al., 1999

Leonard KS, McCubbin D, Blowers P, Taylor BR. Dissolved plutonium and americium in surface waters of the Irish Sea, 1973-1996. *Journal of Environmental Radioactivity* 1999; 44:129-158.

Mobbs and Salbu, 2002

Mobbs HJ, Salbu B. Temporal changes in Technetium-99 activity in algae from the Norwegian coast. Poster. In: Strand P, Børretzen P, Jølle T, eds. International conference on radioactivity in the environment, Monaco 2002. Extended abstracts. Østerås: Norwegian Radiation Protection Authority, 2002. CD-Rom in back of the printed "Extended abstracts".

Moore, et al., 1980

Moore S.M., Feely H.W, Li Y-H. (1980). Radium in sub-arctic waters. *Earth and Planetary Science Letters*, 49, pp 329-340.

NRPA, 2004.

NRPA. Norwegian Radiation Protection Authority. Radioactivity in the marine environment 2002. Results from the Norwegian National Monitoring Programme (RAME). StrålevernRapport 2004:10. Østerås: Norwegian Radiation Protection Authority, 2004.

[http://www.nrpa.no/dokumentarkiv/StralevernRapport10\\_2004.pdf](http://www.nrpa.no/dokumentarkiv/StralevernRapport10_2004.pdf) (06.01.06)

NRPA, 2005a

NRPA. Natural radioactivity in produced water from the Norwegian oil and gas industry in 2003. StrålevernRapport 2005:2. Østerås: Norwegian Radiation Protection Authority, 2005.

[http://www.nrpa.no/dokumentarkiv/StralevernRapport2\\_2005.pdf](http://www.nrpa.no/dokumentarkiv/StralevernRapport2_2005.pdf)

NRPA, 2005b

NRPA. Norwegian Radiation Protection Authority. Radioactivity in the Marine Environment 2003. Results from the Norwegian

National Monitoring Programme (RAME). StrålevernRapport 2005:20. Østerås: Norwegian Radiation Protection Authority, 2005.

OLF, 2005

OLF (Oljeindustriens Landsforening, Stavanger). Miljørapport 2004. Utslippsdata fra de enkelte innterningene på norsk sokkel, se nettsiden til Miljørapport 2004: <http://www.olf.no/miljo/miljorapporter/?27222> (18.09.06)

OSPAR, 2006

OSPAR Commission. Liquid discharges from nuclear installations in 2004. Radioactive substances series, Publication number 297/2006. London: OSPAR, 2006.

Povinec et al., 2003

Povinec PP, Bailly du Bois P, Kershaw PJ, Nies H, Scotto P. Temporal and spatial trends in the distribution of  $^{137}\text{Cs}$  in surface waters of Northern European Seas – a record of 40 years of investigations. *Deep-Sea Research II* 2003; 50: 2785-2801.

Roos et al., 1993

Roos P, Holm E, Thornberg C, Mattsson S. Long-term trend of  $^{239+240}\text{Pu}$  and  $^{99}\text{Tc}$  in brown algae from the Swedish west coast. In: Winter M, Wicke A, eds. Umweltradioaktivität: Radioökologie: Strahlenwirkungen. Köln: TÜV Rheinland, 1993: 663-666.

Roos et al., 1994

Roos P, Holm E, Persson RBR. Comparison of AMP precipitate method and impregnated  $\text{Cu}_2[\text{Fe}(\text{CN})_6]$  filters for the determination of radiocesium concentrations in natural waters. *Nuclear Instruments and Methods in Physics Research A* 1994; 339: 282-286.

Rudjord et al., 2001

Rudjord AL, Føyn L, Brungot AL, Kolstad AK, Helldal HE, Brown J, Iosjpe M, Christensen G. Radioactivity in the marine environment 1999. StrålevernRapport 2001:9. Østerås: Norwegian Radiation Protection Authority, 2001.

[http://www.nrpa.no/dokumentarkiv/StraalevernRapport9\\_2001.pdf](http://www.nrpa.no/dokumentarkiv/StraalevernRapport9_2001.pdf) (06.01.06)

Smith, 1987

Smith AL. Radioactive scale formation, Offshore Technology Conference, Journal of Petroleum Technology, June 1987.

Smith et al., 1998

Smith DL, Knowles JF, Winpenny K. The accumulation, retention and distribution of  $^{95m}\text{Tc}$  in crab (*Cancer pagurus L*) and lobster (*Homarus gamarus L*). A comparative study. Journal of Environmental Radioactivity 1998; 40: 113-135.

Smith et al., 2001

Smith V, Fegan M, Pollard D, Long S, Hayden E, Ryan P. Technetium-99 in the Irish marine environment. Journal of Environmental Radioactivity 2001; 56: 269-284.

Strand et al., 1997

Strand T, Lysebo I, Kristensen D, Birovljev A. Deposits of naturally occurring radioactivity in production of oil and natural gas. StrålevernRapport 1997:1. Østerås: Norwegian Radiation Protection Authority, 1997. (In Norwegian)  
.http://www.nrpa.no/dokumentarkiv/StraalevernRapport1\_1997.pdf (06.01.06)

Strand, 1998

Strand P, Balonov M, Aarkrog A, Bewers MJ, Howard B, Salo A, Tsaturov YS. Radioactivity. In: AMAP assessment report: Arctic pollution issues, Chapter 8. Oslo: Arctic Monitoring and Assessment Programme, AMAP 1998: 525-620. Also available as offprint from AMAP secretariat, Oslo.

Swift, 1985

Swift D. J. The accumulation of  $^{95m}\text{Tc}$  from sea water by juvenile lobsters (*Homarus gammarus*). Journal of Environmental Radioactivity 1985; 2: 229-243.

Topcuoglu and Fowler, 1984

Topcuoglu S, Fowler SW. Factors affecting the biokinetics of technetium ( $^{95m}\text{Tc}$ ) in marine

macroalgae. Marine Environmental Research 1984; 12: 25-43.

UNSCEAR (2000a)

UNSCEAR. United Nations Scientific Committee on the Effects on Atomic Radiation. Sources and effects of ionizing radiation: United Nations report to the general assembly, with scientific annexes. Volume I: Sources. New York: United Nations, 2000.

UNSCEAR (2000b)

UNSCEAR. United Nations Scientific Committee on the Effects on Atomic Radiation, Sources and effects of ionizing radiation: United Nations report to the general assembly, with scientific annexes. Volume II: Effects. New York: United Nations, 2000.

Vakulovsky et al., 1995

Vakulovsky SM, Kryshsev II, Nikitin AI. Radioactive contamination of the Yenisei River. Journal of Environmental Radioactivity 1995; 29: 225-236.

Varskog et al., 1997

Varskog P, Bjerck TO, Ruud AB. Ph-control EDTA titration as an alternative method for the determination of the chemical yield of yttrium in  $^{90}\text{Sr}$  analysis. In: Rapid radioactivity measurements in emergency and routine situations. Teddington: National Physical Laboratory, NPL, 1997: 237-241.

Vartti et al., 2006

Vartti V-P, Illus E, Mattila J, Klemola S, Ikäheimonen TK. Monitoring of radionuclides in the Baltic Sea in 2005. Helsinki Commission. Project Group for Monitoring of Radioactive Substances in the Baltic Sea (MORS), Eleventh meeting, Monaco 2006. HELCOM MORS-PRO 11/2006, Document 3/3. Helsinki: Radiation and Nuclear Safety Authority, STUK, 2006.  
[http://sea.helcom.fi:15037/dps/docs/documents/Monitoring%20and%20Assessment%20Group%20\(MONAS\)/MORS%20PRO%2011,%20202006/33.pdf#search=%22Monitoring%20of%20radionuclides%20in%20the%20Baltic%20Sea%20in%202005%22](http://sea.helcom.fi:15037/dps/docs/documents/Monitoring%20and%20Assessment%20Group%20(MONAS)/MORS%20PRO%2011,%20202006/33.pdf#search=%22Monitoring%20of%20radionuclides%20in%20the%20Baltic%20Sea%20in%202005%22) (18.09.06)

## Appendix

### Analytical methods

During recent years, laboratories at the Norwegian Radiation Protection Authority, and the Institute of Marine Research have both been accredited according to the requirements of NS-EN ISO/IEC 17025. Laboratories and organisations that are not named here have not been accredited according to the requirements of NS-EN ISO/IEC 17025. The scope and conditions governing the accreditation of the institutes mentioned above are quite different, but they have all been accredited for gamma spectrometric measurements. For example, the NRPA is accredited for gamma spectrometric measurements in the energy interval 100-1800 keV (except  $^{226}\text{Ra}$ ), while the other organisations are accredited for gamma spectrometric measurements of  $^{137}\text{Cs}$ . The analytical techniques employed at each institution are described below. The results in this report are not claimed to be accredited results as not all results are accredited.

### The Norwegian Radiation Protection Authority (NRPA)

NRPA analyses alpha, beta and gamma emitters, using the procedures described in this Appendix.

#### *Determination of $^{238}\text{Pu}$ , $^{239+240}\text{Pu}$ and $^{241}\text{Am}$ activity*

The concentrations of  $^{238}\text{Pu}$ ,  $^{239+240}\text{Pu}$  and  $^{241}\text{Am}$  were measured in samples of 200 litres of seawater or 10-20 g of sediment.  $^{242}\text{Pu}$  and  $^{243}\text{Am}$  were added for chemical yield determination. Precipitation from the water samples was achieved using the analytical procedure described by Chen *et al.*, (1991). Different radiochemical separation techniques were applied to separate plutonium and americium from other nuclides using solvent extraction with 10 % TIOA/xylene solution and ion-exchange chromatography with a BIO-RAD AG1-X4 (100-200 mesh) column. Purified americium and plutonium fractions were electrodeposited on stainless steel discs and the activity measured

in semiconductor silicon detectors. As alpha spectrometry is not able to distinguish between  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$ , because the energies of their emitted alpha particles are too close to be resolved, these isotopes are measured and reported as the sum  $^{239+240}\text{Pu}$ . Relative efficiencies of the detectors were in the range 25 to 30 %. The resolution of the detectors, the full width at half maximum (FWHM), was approximately 20 keV at 5486 keV ( $^{241}\text{Am}$ ). Chemical yields obtained from the  $^{242}\text{Pu}$  and  $^{243}\text{Am}$  yield monitor were in the range 40 % to 75 %.

#### *Determination of $^{99}\text{Tc}$ activity*

To determine the activity concentration of  $^{99}\text{Tc}$  in seawater, samples of 50 litres were filtered through a 1-micron polypropylene cartridge to remove suspended particulate matter. Seaweed and other biota samples were dried, milled and homogenized. A 10-20 g dried sample was transferred to a specially designed bottle and carbonised and then dissolved by adding concentrated  $\text{H}_2\text{SO}_4$  followed by  $\text{HNO}_3$ .  $^{99\text{m}}\text{Tc}$  was added to all samples for chemical recovery determination.



**Figure A.1.** Fifty litres of seawater is passed through an ion-exchange column.

The analytical procedure used is a modified version of that developed by Risø National Laboratory, Denmark (Chen *et al.*, 2001). The procedure is outlined briefly below. Technetium is initially separated from the matrix by ion-exchange chromatography using a BIO-RAD AG1-X4 (100-200 mesh) column (Figure A.1) and separation techniques such as precipitation and solvent extraction are then

applied before the technetium is electro-deposited onto stainless steel discs (Figure A.2). The chemical yields were determined by gamma counting of the  $^{99m}\text{Tc}$  tracer in a NaI well-type detector. Typically, the radiochemical yields varied between 70 % and 85 %. After one week, the  $^{99}\text{Tc}$  activity was measured using a low-background anti-coincidence beta counter (Model Risø GM-25-5).



**Figure A.2.** Dried sample material in different containers ready for gamma measurements. To the right,  $^{99}\text{Tc}$  preparations ready for counting.

The limits of detection for 10 g seaweed and 50 l seawater have been calculated to be approximately  $0.5 \text{ Bq kg}^{-1}$  (d.w.) and  $0.10 \text{ Bq m}^{-3}$ , respectively. The limit of detection may vary slightly owing to variations in chemical yield, counting efficiency and the mass of the sample. The total uncertainty in  $^{99}\text{Tc}$  analysis is normally around 10 %.

#### *Detection of gamma emitters*

At NRPA analyses of gamma-emitting nuclides are performed with HPGe detectors. All the detectors are situated in a low-background laboratory to ensure low detection limits. The HPGe detectors have relative efficiencies in the range of 23 % to 40 %. The resolution of the detectors, the full width at half maximum (FWHM) at 1332 keV, was less than 1.9 keV. Three of the detectors cover the energy interval 50-2000 keV, and two cover the interval 20-2000 keV.

Caesium-absorbing filters (sometimes also the prefilter) from the filtering system for seawater samples were dried separately at  $105^\circ\text{C}$  and ashed at  $450^\circ\text{C}$  before the activity was

determined with an HPGe detector. The counting time varied from 1 to 4 days.

Samples of seaweed were dried at  $105^\circ\text{C}$  and homogenized, and placed in containers prior to gamma counting. Sediment samples were freeze-dried and placed in containers prior to gamma counting. Samples were counted for a minimum of 2 days. At NRPA, the detection limits for  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  in the sediment samples ranged from approximately  $0.3\text{-}2 \text{ Bq kg}^{-1}$  (d.w.) Uncertainties in the analysis include uncertainties from counting statistics, calibration and sample preparation and are normally in the range 5% to 20%.

### **The Institute of Marine Research (IMR)**

#### *Detection of gamma emitters*

Sediment samples were transported deep-frozen to IMR, where they were subsequently ground, freeze-dried, homogenized and transferred to polyethylene containers of appropriate size prior to analysis. The gamma counting for  $^{137}\text{Cs}$  detection was performed in IMR's low-background laboratory using a HPGe detector with 30 % relative efficiency and an HPGe detector with 60 % relative efficiency. Both detectors have electric cryostat cooling systems, and 10 cm lead shielding.

### **Institute for Energy Technology (IFE)**

#### *Determination of $^{99}\text{Tc}$*

Samples of dried and homogenized *Fucus vesiculosus* were analysed with regard to  $^{99}\text{Tc}$  content. The pretreatment was performed at Kjeller and the analysis at the Department of Radiation Physics, at Lund University. The following analytical procedure was used at the Department of Radiation Physics at Lund University, Sweden. Technetium was extracted into tri-butyl-phosphate (TBP) from sulphuric acid-hydrogen fluoride solution. Technetium was then back-extracted from the organic phase with a sodium hydroxide solution from which the technetium was electrodeposited onto stainless steel discs, and  $^{99m}\text{Tc}$  was used as a

radiochemical yield determinant. After decay of the yield determinant,  $^{99}\text{Tc}$  was measured with an anti-coincidence-shielded GM counter (Holm *et al.*, 1984).

#### *Determination of $^{90}\text{Sr}$*

For  $^{90}\text{Sr}$ , the standard method using fuming nitric acid was used. The recovery of  $^{90}\text{Sr}$  in the analytical process was monitored by adding  $^{85}\text{Sr}$  as a yield determinant, and the recovery of the daughter nuclide  $^{90}\text{Y}$  was determined by titration with EDTA (Varskog *et al.*, 1997). Finally,  $^{90}\text{Y}$  was measured with an anti-coincidence-shielded GM counter.

#### *Detection of gamma emitters*

At IFE analyses of gamma-emitting nuclides are performed with low-background HPGe detectors. Samples of seaweed were dried at  $105^\circ\text{C}$  and then homogenized, and placed in suitable containers prior to gamma counting.

#### *Determination of $^{239+240}\text{Pu}$*

Samples of dried and homogenized *Fucus vesiculosus* were ashed and treated with *aqua regia* before separation of plutonium isotopes.  $^{242}\text{Pu}$  was used as a tracer for radiochemical yield determination. The separation process involved solvent extraction with 10 % TIOA/Xylene followed by anion exchange on AG1-X4 columns. The plutonium fraction were then electrodeposited on a stainless steel disc and counted using alpha spectrometry.

### **Risø National Laboratory**

#### *Determination of $^{90}\text{Sr}$ in seawater*

Sr was precipitated as strontium carbonate from the seawater. The carbonate was later dissolved in 4 M  $\text{HNO}_3$  and Ca was separated by precipitating Sr and Ba nitrates by adding concentration nitric acid and finally fuming nitric acid. Ba was removed by precipitation of  $\text{BaSO}_4$  followed by a  $\text{Fe}(\text{OH})_3$  to remove yttrium. After a known amount of yttrium carrier had been added, the sample was stored for three weeks to ensure secular equilibrium between  $^{90}\text{Sr}$  and  $^{90}\text{Y}$ . Yttrium was separated by precipitation of  $\text{Y}(\text{OH})_3$  and the activity

measured by beta counting. The  $\text{Y}(\text{OH})_3$  precipitate was then dissolved in 6 M  $\text{HNO}_3$  and yttrium was precipitated as an oxalate by adding  $\text{H}_2\text{C}_2\text{O}_4$  to the solution. After the precipitate had been filtered from the solution and dried, the recovery of yttrium was measured by weighing the oxalate precipitate. The recovery of Sr was determined by using  $^{85}\text{Sr}$  as a radiochemical tracer.

#### *Determination of $^{226}\text{Ra}$ and $^{228}\text{Ra}$ in seawater*

Radium was coprecipitated with  $\text{MnO}_2$  from 10 l of seawater, after  $^{133}\text{Ba}$  had been added as a yield determinant. The  $\text{MnO}_2$  precipitate was then dissolved in hydrochloric acid and hydrogen peroxide, and after the addition of sulphuric acid,  $\text{K}_2\text{SO}_4$  and  $\text{Pb}(\text{NO}_3)_2$ , radium was coprecipitated with  $\text{PbSO}_4$ . The precipitate was dissolved in EDTA at  $\text{pH} = 10$ , and transferred to a liquid scintillation vial. After measuring the radiochemical yield with gamma spectrometry, a liquid scintillation cocktail was added (OptiFluor O). The sample was then stored for about four weeks after which the activity of  $^{226}\text{Ra}$  was measured through  $^{214}\text{Po}$  with a low-background, liquid scintillation counter (Quantulus). The laboratory work was carried out at Risø National Laboratory. The  $^{228}\text{Ra}/^{226}\text{Ra}$  ratio was determined by pumping several hundred litres of seawater through a  $\text{MnO}_2$ -impregnated polyethylene filter. The filter was later ashed and  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  were analysed by gamma spectrometry.

### **The Norwegian University of Life Sciences (UMB)**

#### *Determination of $^{99}\text{Tc}$*

Blades and stipes were separated, dried and ground before analysis. Fish was freeze dried, ashed and ground prior to analysis.

Tc-99 was extracted using microwave acid digestion followed by separation on a TEVA ion-exchange column. Tc-99m was used as yield monitor for the extraction. The concentration of Tc-99 in the extracts was determined using Inductively Coupled Plasma - Mass Spectrometry (ICP-MS).



**StrålevernRapport 2006:1**  
Virksomhetsplan 2006

**StrålevernRapport 2006:2**  
Statens strålevern i Mammografiprogrammet. Resultater fra teknisk kvalitetskontroll hentet fra databaseprogrammet TKK

**StrålevernRapport 2006:3**  
Avvikshåndtering ved norske stråleterapisentre

**StrålevernRapport 2006:4**  
The Norwegian UV Monitoring Network 1995/96 - 2004

**StrålevernRapport 2006:5**  
Sikkerhet ved russiske RBMK-reaktorer  
En oppdatert gjennomgang av status

**StrålevernRapport 2006:6**  
Radiologi i Noreg. Undersøkningsfrekvens per 2002, tidstrender, geografisk variasjon og befolkningsdose

**StrålevernRapport 2006:7**  
Tiltak mot radon i privatboliger  
Oppsummering av tiltak under Nasjonal kreftplan 1999-2003

**StrålevernRapport 2006:8**  
K-159. Havariet av den russiske atombåten K-159 og den norske atomberedskapsorganisasjonens håndtering av ulykken

**StrålevernRapport 2006:9**  
Monte Carlo Simulations for Gamma Measurements in Monitoring and Emergency Situations

**StrålevernRapport 2006:10**  
Terrestrial Monitoring in Øvre Dividalen

**StrålevernRapport 2006:11**  
Virksomhetsrapport for norske stråleterapisentre 2003-2004

**StrålevernRapport 2006:12**  
Gammaspectrometriske flymålinger og radon

**StrålevernRapport 2006:13**  
Kvalitetskontroll av ikke-dosimetriske parametre ved CT-basert planlegging av stråleterapi